



DEPARTMENT OF PHYSIOLOGY  
COLLEGE OF MEDICINE  
UNIVERSITY OF FLORIDA  
GAINESVILLE, FLORIDA 32610

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Box J-274  
J. HILLIS MILLER HEALTH CENTER  
Phone: 904-392-3791

August 15, 1989

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Dr. Peter Kent  
Research and Development Command  
Naval Medical Command  
Bethesda, MD 20814-5044

RE: 00014-86-K-0123

Dear Dr. Kent:

This is the final technical report on this contract. It fulfills the requirement of the contract dated Jan. 1, 1986, page 2.

I) Summary of Work

- I. This project was designed to evaluate the feasibility of using High Frequency Ventilation (HFV) or High Frequency Oscillations (HFO) in deep sea diving. This novel method of ventilation has raised great expectations among clinicians since its introduction in the early 1970s. The technique has found well-defined applications in intensive care and in operative anesthesia. It was justified to ask if the technique could be used in deep sea diving.
- II. Several series of dog experiments were performed to test in the laboratory the feasibility and the efficiency of using HFV with a gas mixture that simulates the conditions in deep sea diving. The main result is that HFV may be used with the same efficiency as at sea level. The technique was used for up to 4 hours.

There is a notable disadvantage in the use of HFV as compared to conventional techniques. HFV results in high pressures in the airways which could potentially cause injury to the inner lining of the airways.

The decision if and when HFV should be used in deep sea diving is only partially answered by our results. The contract was to conduct a feasibility study and our study shows that HFV can indeed be used. No application study was conducted. For details, see report A.

- III. Experiments were completed to test whether helium (which is used extensively in deep sea diving) has a specific, favorable or unfavorable, effect on respiration if HFV is used. No such effect was found. For details, see report A.
- IV. Model experiments were performed using a number of models simulating the different bronchi found in the airways. These experiments had the purpose of documenting theoretically the results obtained in dogs. It was found that the increase of density that results from deep sea diving has opposite effects in large tubes such as the trachea

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Dr. Peter Kent  
August 15, 1989  
Page Two

as compared to small tubes such as the small airways. The fact that the effects are opposite explains why the efficiency of ventilation is not affected in deep sea diving; the effects, while notable within subsections of the airways, cancel in the overall result. For details, see report B.

- V. We studied in model experiments whether the uptake of  $O_2$  and the elimination of  $CO_2$  are affected differently by HFV. We found no difference.
- VI. We were quite interested in the finding mentioned in paragraph V. Our theory would have led us to believe that there should be a small difference. This difference would be of little clinical importance, but we were interested in it anyway.

The result of this interest was the invention of a new, promising technique to separate gases. This technique has been patented (#4,770,675). For details, see report C. It is a technique that might be of use to the Navy for the recovering of helium used in deep sea diving.

2) List of Technical Reports

- ↓ Report A: High frequency ventilation in dogs with three gases of different densities.
- ↓ Report B: Dispersion of gases by high frequency oscillation of tube systems of different sizes and with different gas media: model experiments.
- ↓ Report C: Diffusional separation of gases and solutes in oscillatory flow.
- ↓ Report D: Patent #4,770,675.

Copies of the technical reports are included.

- 3) Two of the above technical reports (A and B) have been submitted for publication. C is being prepared for publication.

Sincerely,

*Marc Jaeger*

Marc Jaeger, M.D.  
Professor

MJ:krf

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HIGH-FREQUENCY VENTILATION IN DOGS WITH THREE  
GASES OF DIFFERENT DENSITIES

Marc J. Jaeger  
Dept. of Physiology  
College of Medicine  
University of Florida

Address:  
Dept. of Physiology  
College of Medicine  
Box J-274, JHMHC  
University of Florida  
Gainesville, FL 32610

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### ABSTRACT

Dogs were ventilated with a high frequency oscillation, HFO, device varying the frequency (2 - 20 Hz), the tidal volume (25 - 100 ml), and the resident gas (He, N<sub>2</sub>, SF<sub>6</sub>). Tidal volume was measured with a body plethysmograph. Blood gases were measured after a quasi steady state was established. The kinematic viscosity of the breathing gas mixture, which changed by 1700%, was found to have little effect on arterial P<sub>O2</sub> and P<sub>CO2</sub>. The results are consistent with findings in a model which consisted of tubes of different diameters and with the theory of Taylor-type diffusion. In addition, experiments were performed reducing and increasing the equipment dead space. This resulted in changes of P<sub>O2</sub> and P<sub>CO2</sub> that were appreciably less than those resulting from variations of tidal volume of the same magnitude. These results suggest that, high frequency ventilation, HFV, at increased and decreased ambient pressure is technically possible.

#### Key Words:

Gas exchange, high frequency ventilation; high frequency oscillations; augmented diffusion; deep sea diving, gas separation.

## INTRODUCTION

High frequency oscillation ventilation (HFOV) has been found to be of use in some well-defined clinical conditions (4). It has also been a great scientific stimulus since it contradicts an old physiological dogma which states that tidal volume must exceed dead space for gas exchange to occur. This paper investigates the effect of resident gas density on the gas exchange in dogs. We replaced  $N_2$  in air either with He or with  $SF_6$ . Our hypothesis is that a change of the resident gas would bring with it a change of gas exchange. We wish to evaluate possible advantages and disadvantages of the use of resident gases with different densities. We also evaluate possible use of HFOV at increased or decreased ambient pressure. Finally, we also undertook these experiments to get additional insight into the mechanism of HFOV.

Several mechanisms are thought to contribute to the gas transport in HFOV (1). Presumably the most important is Taylor-type dispersion. It is also the best understood. But other mechanisms such as direct ventilation of hilus-nigh alveoli (1), convective dispersion due to disturbances of the flow at branchings (6), and Pendelluft also play a role (8). Taylor-type dispersion is a form of diffusion and is directly dependent on the molecular diffusion coefficient  $D_m$ . Since  $D_m$  of the diffusing gas ( $CO_2$  or  $O_2$ ) is strongly dependent on the molecular weight of the resident gas (in our case He,  $N_2$ , or  $SF_6$ ), we hypothesized that a change of the resident gas would have a noticeable effect on gas transport.

## METHODS

Fourteen mongrel dogs were ventilated at several breathing rates and tidal volumes with a high-frequency oscillation device using three different gas mixtures: He/O<sub>2</sub>, air and SF<sub>6</sub>/O<sub>2</sub>. Each condition was maintained for 12 minutes. Afterwards, arterial blood was withdrawn and an alveolar gas sample was obtained and analyzed for CO<sub>2</sub> and O<sub>2</sub>. Experiments were also performed with an extended and shortened airway. The procedures and methods used in this study were approved by the institutional review board.

The dogs (average weight: 16.2 ± 1.6 kg) were anesthetized with pentobarbital (30 mg/kg) and intubated with a multiple-lumen, 0.8 cm ID, cuffed endotracheal tube. Femoral artery and vein were cannulated and the dog was placed in a plethysmograph. After stabilizing the dog with CMV with a frequency ( $f$ ) of 8 bpm and a tidal volume ( $V_T$ ) of about 300 ml, blood gases were obtained. The first HFO experiment was usually done on air with a  $f = 10$  Hz and  $V_T = 50$  ml. Afterwards, conditions ( $f$ ,  $V_T$ , gas composition) were varied at random. After completing a measurement at a particular condition, the dog was inflated to near TLC with a large syringe and ventilated with air for five minutes using CMV and a Harvard pump. Each dog was exposed to each condition of a particular series so that graphs shown below always imply repeated measurements in the same animals.

Blood gases were measured with a Radiometer (BMS 3 Mk 2) blood gas machine which was repeatedly calibrated and held at 37°C. The dogs were maintained at the same temperature. The plethysmograph was opened for each blood sampling and the sample was taken directly from the cannula.

The complete setup shown schematically in Fig. 1 included: a variable frequency and variable stroke volume pump (#1; linear motor; amplifier designed by Dr. John Lehr, Harvard School of Public Health, Boston, MA) and the constant volume body plethysmograph (#2). The pressure in the plethysmograph was sensed by a Datametrics Barocel pressure transducer

(#3; Gould), the output of which was monitored on an oscilloscope. The box was calibrated daily with the dog inside and tested for leaks. The frequency response was tested by connecting the pump directly to the plethysmograph and found to be within  $\pm 5\%$  up to 50 Hz with  $V_T = 80$  ml. The volume displacement measured by the plethysmograph was used to determine  $V_T$ . We made measurements at  $V_T = 25, 50, 75$ , and 100 ml with a frequency of either 5, 10, or 15 Hz. The body plethysmograph senses any volume displacement within the chest. Most of this displacement is due to a change of alveolar volume. Changes of volumes of the conducting airways, however, are also sensed (5). The amount of volume change of the dead space is difficult to evaluate and may be appreciable when the pressure amplitude in the airways is high (see below). We assumed for clarity that the measured volume displacement in the plethysmograph was due to alveolar volume changes and did not apply any correction.

Blood pressure was continuously monitored with transducer #4. Humidified fresh gas was supplied (#5) at a rate of 20 L/min either from an air source or from a Linde mass gas controller that provided either 79% He with 21%  $O_2$  or 79%  $SF_6$  with 21%  $O_2$ . Port #6 was used to measure the pressure at the airway opening ( $P_{ao}$ ); it was located 15 cm from the X-piece #7. This port was also used to monitor at intervals  $F_{O_2}$  and  $F_{CO_2}$  of the inspired gas. An end-tidal sample was obtained at the end of each experiment by inflating the dog with a large syringe with a  $V_T$  of 200 ml and removing the same volume again; it was used to assess alveolar  $F_{O_2}$  and  $F_{CO_2}$  using gas sampled at the tip of the ET-tube (#8). No correction was made for dilution. The X-piece #7 had an ID of 1/2" and was symmetrical.

The bias tube (#9) was 8 meters long and 1.59 cm in ID. It was found that the gas in the X-piece (#7) was diluted by air if the tube was shorter than 8 m. This dilution was undesirable when the dog was ventilated with  $He/O_2$  or  $SF_6/O_2$ .

The pressure drop in the intubation system from port #6 to the tip of the ET-tube was measured in a separate experiment in which the dog was replaced by a 2-liter rubber bag. At 10 Hz and with a  $V_T$  of 50 ml, e.g., it was 13.5, 24, and 92 cm  $H_2O$ , for  $He/O_2$ , air, and  $SF_6/O_2$ , respectively. The average pressure in the bag, representing, presumably, average intratracheal pressure determining lung volume, was 0.7, 1.2, and 1.6 cm  $H_2O$  at the same conditions. This nominal average pressure prevented changes of functional residual volume due to airway pressure (12). The pressure amplitude in the trachea, determined by subtracting the pressure drop in the ET-tube from measurements made at the mouth opening, was determined at  $f = 10$  Hz and  $V_T = 50$  ml. The average in 8 dogs was  $35.0 \pm 9.7$ ,  $104.3 \pm 51.5$  and  $153.4 \pm 113.9$  cm  $H_2O$  for  $He/O_2$ , air, and  $SF_6/O_2$ , respectively.

Two further series of measurements were made on six of the same dogs. The equipment dead space (i.e., the space between the ET-tube and the X-piece #7) was either increased by 40 ml or decreased by injecting the bias flow into the trachea via one of the lumens of the ET-tube. The injecting, presumably, reduced the  $V_D$  of the animal by the volume of the ET-tube up to the X-piece. We estimate this reduction to be about 40 ml.

A separate series of experiments were done on 8 additional mongrel dogs. In these experiments, ventilation with the three gas mixtures was maintained for 4 hours. The frequency was either 10 or 15 Hz and the tidal volume adjusted for normocarbica. These tests were done to evaluate the effect of the different gas mixtures on longer term ventilatory and circulatory function. The tracheas of these dogs were examined histologically after completion of each experiment.

The physical characteristics of the three gas mixtures used are listed in Table 1. It may be seen that the molecular diffusion coefficients of  $O_2$  and  $CO_2$  are about 4 to 5 times higher in  $He/O_2$  than in  $SF_6/O_2$ . From this, one would expect a priori large effects of changes of the resident gas on gas exchange.



The statistical analysis was performed with a 2-way analysis of variance which analyzed the blood gas data as a function of  $f$  and gas composition or  $V_T$  and gas composition.

## RESULTS

The effects of changing the resident gas on  $P_{aCO_2}$  and  $P_{aO_2}$  are shown in Figs. 2 to 5. The comparison is made in two series of experiments; in one (Figs. 2 and 3),  $V_T$  is kept constant at 50 ml while  $f$  is varied from 5 to 15 Hz; in the other,  $f$  is kept constant at 10 Hz while  $V_T$  is varied from 25 to 100 ml (Figs. 4 and 5). It may be seen that in all four panels there is no appreciable difference in the blood gases as a function of resident gas density ( $P>0.05$ ). An exception to that statement has to be made for Fig. 2, where average  $P_{aCO_2}$  with  $SF_6$  as resident gas is slightly higher than with the other two gases ( $P<0.05$ ). But the observation is isolated and we find it justifiable to say that we find no appreciable effect of resident gas density on blood gases. The four panels show other effects as well.  $P_{aCO_2}$  decreases with increasing  $f$  and  $V_T$  (Figs. 2 and 4;  $P<0.05$ ) and  $P_{aO_2}$  increases as a function of the same variables (Figs. 3 and 5;  $P<0.05$ ). These changes are expected.

The data shown allow one to compute the alveolo-arterial  $P_{O_2}$  gradient. Since the analysis does not show differences between gas mixtures, we compute the (A-a) gradient averaged for all three gas mixtures: the (A-a) gradient decreases with increasing frequency from 25.0 mm Hg at 5 Hz to 3.6 at 15 Hz ( $P>0.05$ ). The effect is much more pronounced when  $V_T$  is varied: the (A-a) gradient decreases from 54 mm Hg when  $V_T = 25$  ml to 4 mm Hg when  $V_T$  equals 100 ml ( $P<0.05$ ).

End-tidal  $P_{CO_2}$  values closely followed changes of  $P_{aCO_2}$ , but were always slightly higher; they are not shown for brevity's sake.

The effect of injecting the fresh gas at the tip of the ET-tube and the effect of extending the equipment dead space by 40 ml are shown for 6 dogs in Fig. 6. Arterial  $P_{CO_2}$  increases on the average by 5.8 mm Hg when equipment dead space is raised by 40 ml; it decreases by 4.2 mm Hg when fresh gas is injected at the tip of the ET-tube ( $P<0.05$ ). The changes of arterial  $P_{O_2}$  with variations of dead space are inverse to those of  $P_{CO_2}$  ( $P<0.05$ ). The decrease of arterial  $P_{O_2}$  is larger in  $SF_6$  (55 mm Hg) than in He (31 mm Hg).

## DISCUSSION

This study was undertaken to investigate the possibility of using HFOV at increased and at decreased barometric pressure. In a preliminary attempt to answer the question, we studied the effect of changes of gas density by altering the molecular weight of the resident gas using either He, N<sub>2</sub>, or SF<sub>6</sub>. The question whether changes of gas density of the resident gas adequately simulate changes of barometric pressure will be discussed. But if the assumption is valid, an initial result of our study is that, indeed, HFOV can be used at increased or at decreased ambient pressure. We found no notable changes in blood gases when resident gas density was varied over a 17-fold range. This caused a large variation of kinematic viscosity and a fairly large variation of the molecular diffusion coefficient (Table 1).

Robertson et al. published in 1982 a similar study. They fixed the stroke volume at 105 ml and f at 10 Hz. They found P<sub>CO<sub>2</sub></sub> to be 2 mm Hg lower in He than in N<sub>2</sub>. We find a similar small difference.

There is no accepted theory on gas transport in HFOV. The main reason for this lack of understanding is the complexity of the bronchial geometry with frequent branchings, short bronchial length, large variations in bronchial diameter from trachea to respiratory bronchi, and pronounced asymmetry. One possible mechanism is based on this asymmetry: some alveoli are located hilus-nigh and therefore close to the airway opening (1). They may receive bulk ventilation when tidal volume is small, while other alveoli that are more distant receive less ventilation and certainly not by bulk flow. The number of alveoli being ventilated thus increases with V<sub>T</sub>. Our data support this model. The (A-a)<sub>O<sub>2</sub></sub> gradient estimated with the alveolar gas equation decreases from 54 mm Hg to 4 mm Hg when V<sub>T</sub> is raised from 25 to 100 ml, while f = 10 Hz (Figs. 4 and 5). The (A-a)<sub>O<sub>2</sub></sub> gradient varies in the same way with the three gases, supporting the assumption of a bulk flow mechanism. Other mechanisms,

such as Pendelluft, may also explain changes of an  $(A-a)_{O_2}$  gradient. But one would expect Pendelluft to vary with gas density and with inertance (2,3,8).

Another important mechanism in gas transport consists in eddies and differences in the shape of the velocity profile between inspiration and expiration (6). Eddies contribute to radial mixing in the airways. They increase in number and energy with flow rate (i.e. with  $f$  and  $V_T$ ) and with gas density. They diminish when the radius of the bronchus becomes smaller. If these eddies were the sole mechanism of transport, one would expect to see a definite effect of gas density on gas exchange. One would also expect to see a quantitatively similar effect of frequency and tidal volume on gas transport. We find a much larger effect of  $V_T$  (Fig. 2) than of  $f$  (Fig. 3) on  $P_{CO_2}$ . We therefore believe that this mechanism does not explain our observations if taken alone.

Taylor dispersion is the only mechanism that can be analyzed mathematically. The theory was developed by Watson for laminar flow in straight, unbranched tubes (11). It does not apply to branched tubes, but Paloski et al. have shown experimentally that Taylor-type dispersion plays a major role in gas transport in short, branching tubes, too (9). They find a quantitative rather than qualitative difference between branched and unbranched tubes. Taylor dispersion is based on diffusion between the fast-moving core of the gas in the center of a tube and the slow-moving boundary layers. Since it is a diffusion process, the molecular diffusion coefficient plays a major role and one would expect notable changes of gas exchange when this coefficient varies. In our experiments, the coefficient varied over a 4- to 5-fold range (Table 1). The change is due to the variation of the density of the resident gas;  $CO_2$  diffusivity in He is higher than in  $SF_6$ . Thus, one would intuitively expect to see large changes of gas exchange in dogs as a function of resident gas density if Taylor dispersion were the main mode of transport. A closer analysis of the theory, however, proves this expectation wrong.

Watson's theory predicts that the density of the resident gas has opposite effects in small and large tubes: in small tubes the dispersion rises with rising density; in large ones the dispersion falls with density. The effect is sharply dependent on tube diameter and gradually changes when tube diameter increases: when tube diameter is small (ID = 0.1 cm), dispersion is approximately proportional to the square root of density; this relationship weakens with increasing ID until, when the ID is between 0.2 and 0.5 cm, the theory predicts no effect of density at all. As the radius further increases, the relationship changes, and with a diameter of about 2 cm the relationship is approximately proportional to the inverse of the square root of density. We have confirmed experimentally these effects of resident gas density on gas transport in tubes of different diameters; the data are presented in a separate paper together with an analysis of the theory (\_\_\_). We also found that if tubes of different diameters are connected in series, the effects of density are attenuated. With proper choice of tube diameter, almost complete cancelling of the effect of density was found.

The gas transport from airway opening to alveoli may be seen as the sum of the transports in individual generations. If the effect of density is opposite in different generations, attenuation or cancelling will occur. The large range of diameter in the airways promotes such cancelling. By coincidence, the diameter at which the effect of density changes from proportionality to inverse proportionality is located in the middle of the branching system of the airways (generation 4 to 8 in humans; higher in dogs). Attenuation of density effects may also occur when two mechanisms coexist in the same generation. Watson's theory predicts that in large tubes such as the trachea or the ET-tube, dispersion is inversely related to density; yet branching may result in eddies with opposite effects; eddy formation increases with density. Resident gas density thus influences the various mechanisms which promote gas transport in the airways in opposite ways. This may explain why, in dogs, resident gas density has so little effect.

We believe that the experimental data in dogs support the view that several mechanisms contribute to the gas exchange in HFOV. The data do not allow one to determine the magnitude of contribution to gas exchange by the different mechanisms. But one observation may serve as an indicator. We find with others (1) that variation of  $V_T$  has a much larger effect on gas exchange than variation of  $f$  (Figs. 2-5). This is more typical for Taylor dispersion than for eddy formation. We therefore believe with Paloski that Taylor dispersion is the major mechanism even in branched systems such as the airways. The effect of asymmetry of velocity profiles and of asymmetry of alveolar geometry share the role of secondary mechanisms.

In many laws of fluid mechanics, changes of barometric pressure and variations of the molecular weight have the same effect if fluid density varies by the same amount. It is not known if this is true for Watson's theory; the theory was developed for incompressible fluids only. The experimental work performed to confirm the theory was performed in gases with no notable deviations from the theory (for references see companion paper). One may therefore assume that the data obtained with He simulate a reduction of barometric pressure, and that data obtained with  $SF_6$  simulate data at increased pressure. The pressure range corresponding to the properties listed in Table 1 range from 0.3 to 22 ata. Other factors, however, need to be considered. We found the pressure amplitude at the airway opening and in the trachea to be greatly elevated when using  $SF_6$ ; the pressure in the trachea, e.g., varied between + and -76 cm  $H_2O$  with  $SF_6$  when  $V_T$  and  $f$  were chosen to produce eucapnia. Such pressure oscillations could cause mechanical stress to the trachea. We found no signs of histological injury in the mucosa when HFOV was applied for 4 hours. But concern still remains.

Gas compressibility is greatly reduced when ambient pressure rises such as in diving, since compressibility is inversely proportional to ambient pressure. This could increase the

mechanical stress on the trachea in deep diving. At reduced ambient pressure the stress would be lower than at sea level. The increased compressibility at altitude would have another effect: it would reduce tidal volume because of compression and decompression of gas in the airways and in the equipment. This would lead to reduced ventilation if not taken into account by adjusting the stroke volume.

In a separate series of experiments we enlarged and reduced the size of the dead space while keeping  $V_T$  and  $f$  constant (Fig. 6). The purpose was to test the best practical use. The estimated variation of  $V_D$  was 80 ml; it caused a fall of  $P_{O_2}$  by 43 mm Hg (average of 3 gas mixtures) and a rise of  $P_{CO_2}$  of 12 mm Hg. The difference between  $O_2$  and  $CO_2$  is, presumably, related to hilus-nigh alveoli receiving less ventilation when  $V_D$  is enlarged. It is also notable that a change of  $V_D$  by 80 ml causes a smaller variation of  $P_{O_2}$  and  $P_{CO_2}$  than a variation of  $V_T$  by 75 ml (Figs. 4 and 5). This is suggestive of Taylor dispersion for which the theory predicts that ventilation is proportional to  $V_T^2/V_D$  (7). Moreover, the  $O_2$  partial pressure difference in the ET-tube system (obtained by subtracting the  $P_{O_2}$  with enlarged dead space from the value with reduced  $V_D$ ) is smaller in He than in  $SF_6$ . This, as mentioned earlier, is suggestive of Taylor-dispersion. Thus, the experiment of varying the size of  $V_D$  yields a number of indications all pointing to the important role of Taylor dispersion, even in large airways.

### ACKNOWLEDGMENTS

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Table 1  
Physical Gas Properties

	He/O <sub>2</sub>	Air	SF <sub>6</sub> /O <sub>2</sub>	Heliox at 22 ata
Density*, g/L, dry, at 37°C	.303	1.29	4.80	3.05
Kinematic viscosity*, cm <sup>2</sup> /sec	.96	.162	.054	.055
Diffusion coefficient**, cm <sup>2</sup> /sec for CO <sub>2</sub> in	.56	.16	.09	.030
Diffusion coefficient**, cm <sup>2</sup> /sec for O <sub>2</sub> in	.65	.19	.11	.035

\* from Techniques in Life Sciences, ed. A. B. Otis, Elsevier, 1984, p. 413/4.

\*\* Bird, Stewart, and Lightfoot, Transport Phenomena, New York, Wiley, 1960, p. 505.

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### LEGENDS TO THE FIGURES

- FIGURE 1:** Schematic of the experimental setup. 1. Reciprocating pump. 2. Constant volume plethysmograph. 3. Pressure transducer sensing the pressure in the plethysmograph and calibrated for tidal volume. 4. Measurement of arterial vascular pressure. 5. Inflow of bias gas, either He/O<sub>2</sub>, air, or SF<sub>6</sub>/O<sub>2</sub>. 6. Pressure transducer measuring the pressure at the tip of the ET-tube. 7. X-piece. 8. Analyzer sampling gas at the tip of the ET-tube and sensing F<sub>CO2</sub> and F<sub>O2</sub> during end-tidal maneuver. 9. Bias tube.
- FIGURE 2:** Decline of P<sub>aCO2</sub> with increasing f is not significant (P>0.05). The average P<sub>CO2</sub> in SF<sub>6</sub> is higher than in either N<sub>2</sub> or He (P<0.05).
- FIGURE 3:** Increase of P<sub>aO2</sub> with increasing f is statistically significant (P<0.05). There is no difference between gases (P>0.05).
- FIGURE 4:** Decline of P<sub>aCO2</sub> with increasing V<sub>T</sub> is statistically significant (P<0.05). The difference between the three gases is not significant (P>0.05).
- FIGURE 5:** Increase of P<sub>aO2</sub> with increasing V<sub>T</sub> is statistically significant (P<0.05). There is no difference between the three gases (P>0.05).
- FIGURE 6:** Effect of changes of V<sub>D</sub> on P<sub>aO2</sub> and P<sub>aCO2</sub>. The increase of V<sub>D</sub> causes a drop of P<sub>O2</sub> and a rise of P<sub>CO2</sub>. The drop of P<sub>O2</sub> is 30 mm Hg in He and 55 mm Hg in SF<sub>6</sub> (P<0.05).

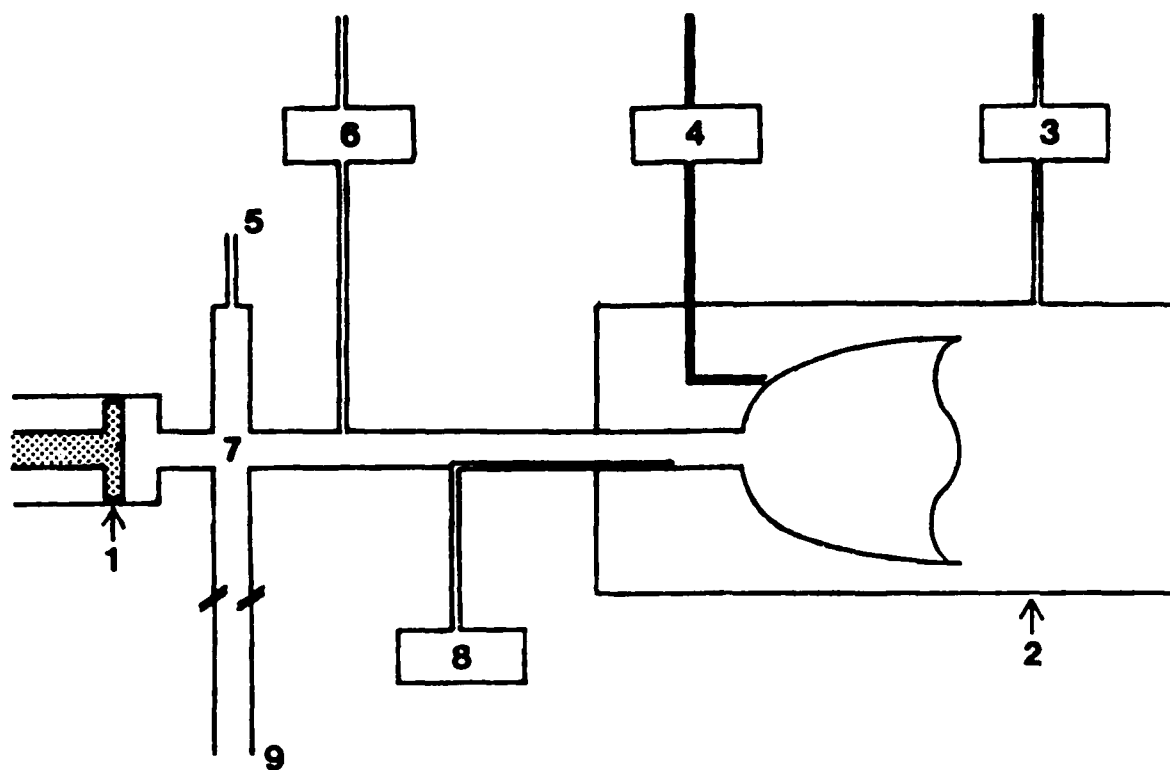


Fig. 1

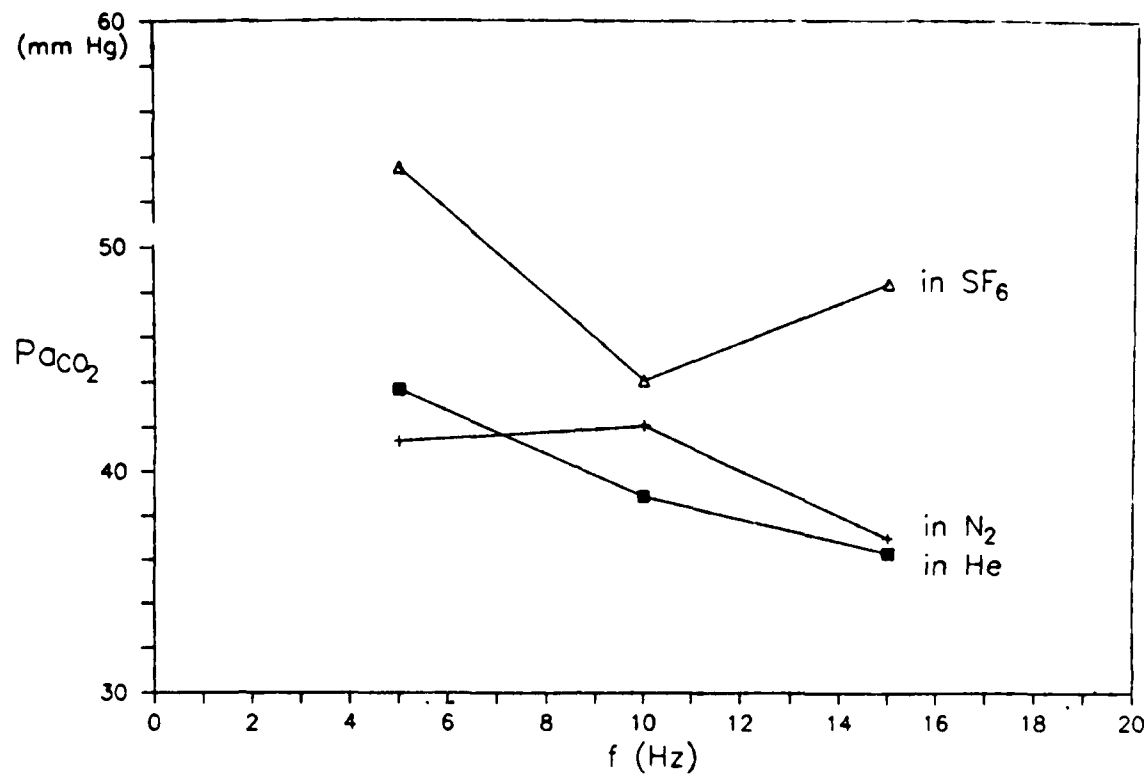


Fig. 2

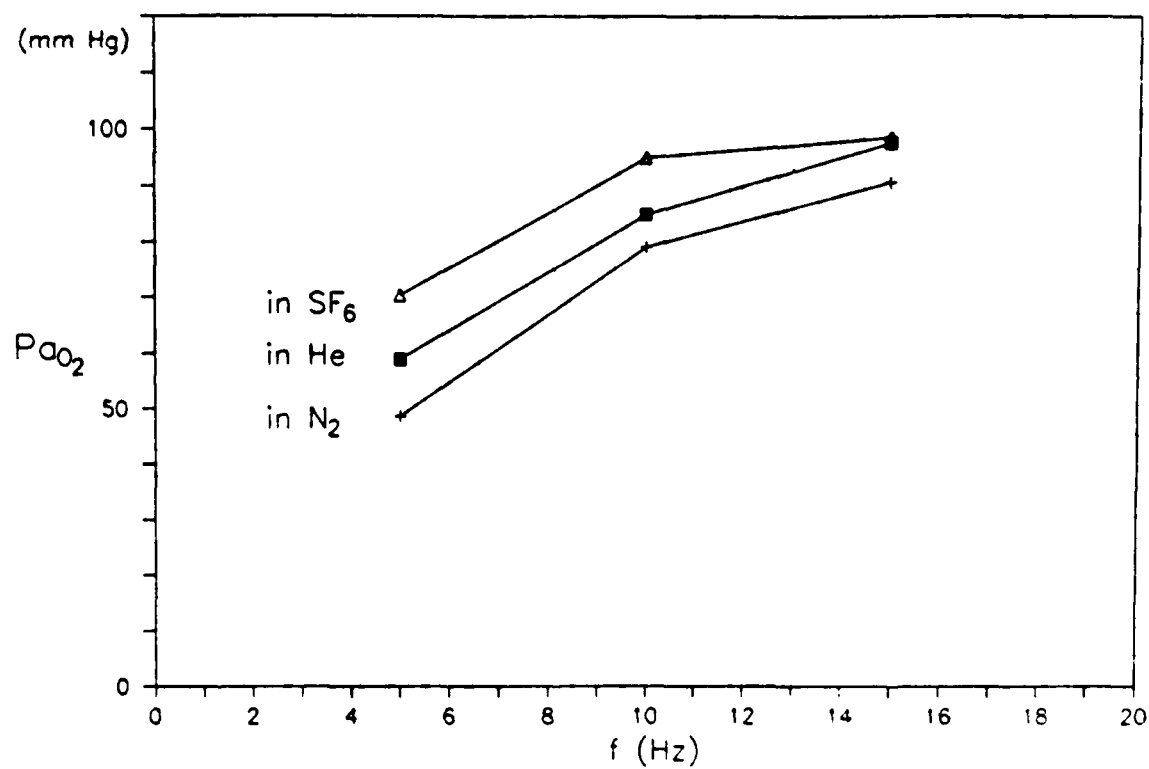


Fig. 3

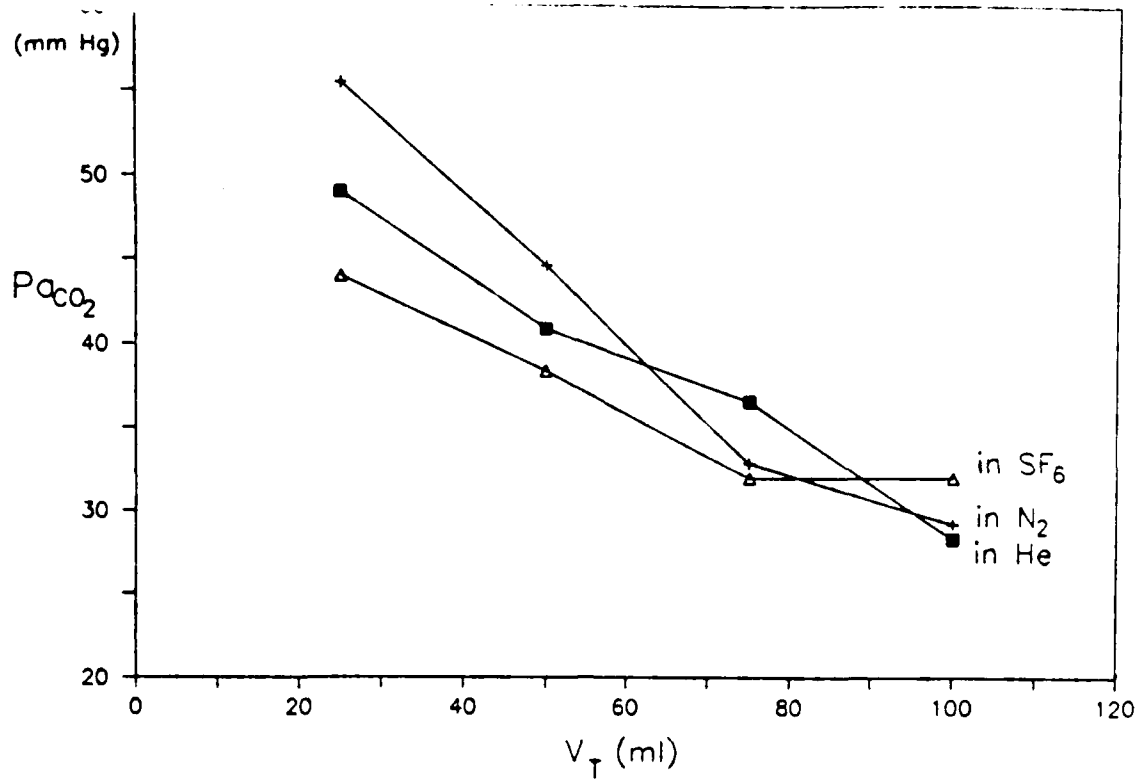


Fig. 4

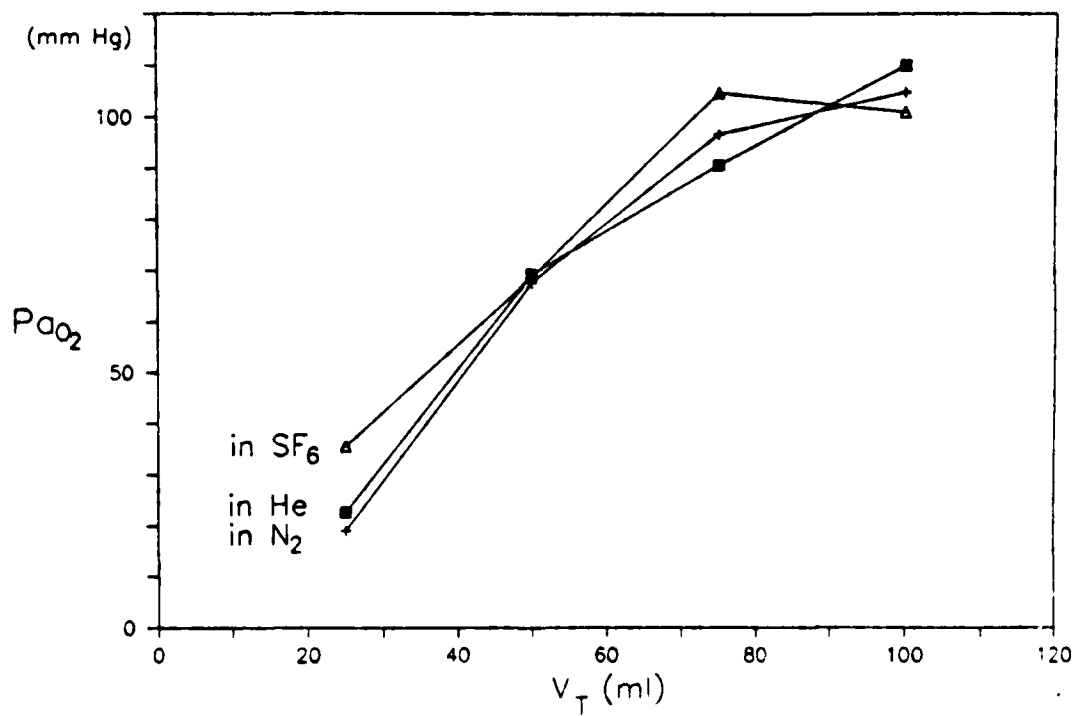


Fig. 5

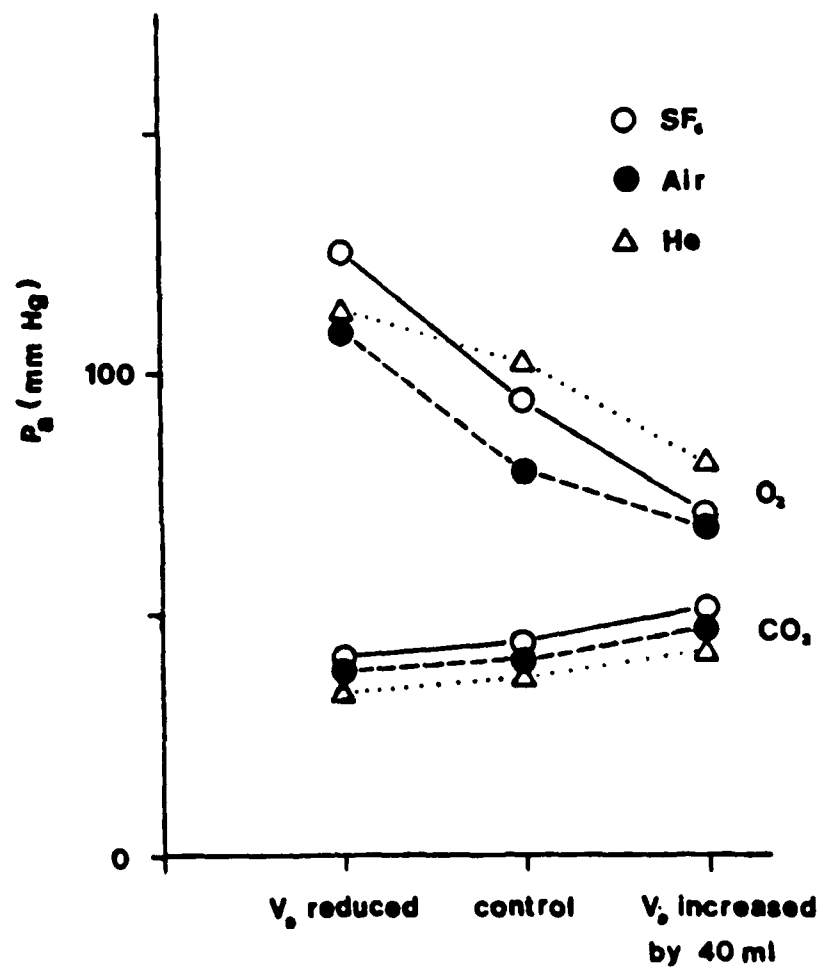


Fig. 6



The dispersion of gases by high frequency oscillation  
in tube systems of different sizes  
and with different gas media: model experiments\*

Marc J. Jaeger

Department of Physiology  
College of Medicine  
University of Florida

Address:

Box J-274

J. H. Miller Health Center

University of Florida

Gainesville, FL 32610

\* supported by U.S. Navy grant N00014-86-K-0123.

## ABSTRACT

This study investigates the effect of variations in the kinematic viscosity of the resident gas on the dispersion of a diffusing gas ( $\text{CO}_2$ ) during high frequency oscillations (HFO). The experiments were conducted in cylindrical tubes with various diameters, in branched tubes, at various frequencies, and at various oscillation amplitudes. Three resident gases were used: He,  $\text{N}_2$ , and  $\text{SF}_6$ . Tube radius was found to be a major factor determining the dispersion of a gas along its concentration gradient in HFO. In large tubes, with a Womersley number  $\alpha$  exceeding 20, the dispersion of a gas increases when the density of the resident gas is reduced. In contrast, in small tubes with  $\alpha$  less than two, the diffusion of a gas increases when the density of the resident gas is raised. In systems which combine tubes of different sizes, the effects cancel. We also discuss, more briefly, the effects of variations of the properties of the diffusing gas on HFO. The effects are opposite in small and large tubes and may also cancel when tubes of different size are connected in series. We propose that these results may apply to a network such as the bronchial tree.

### Key words:

High frequency ventilation, augmented diffusion, gas exchange, gas density.

## INTRODUCTION

The study was planned as a companion to experiments conducted in dogs. We find in dogs that changing the resident gas from  $N_2$  to He or to  $SF_6$  has little effect on the steady-state gas exchange ( $P_{aO_2}$ ,  $P_{aCO_2}$ ) in high frequency oscillation ventilation (HFOV). Almost any fluid mechanical mechanism attempting to explain the gas exchange in HFOV is expected to be influenced by the density of the resident gas, especially if the change of density is large (1). One would expect, e.g., augmented diffusion of  $O_2$  and  $CO_2$  to change greatly if resident gas properties are changed. Yet we do not find such changes in controlled dog experiments. To help elucidate this puzzling contradiction, we study in this paper the effect of Taylor dispersion in straight tubes and compare our data with Watson's theory (15) and with data obtained in branched tubes by Paloski et al. (13). We also discuss the effect of changing the diffusing gas while the resident gas is kept the same. We conclude that the results in dogs are in agreement with theoretical predictions.

## METHODS

The experimental setup was very similar to that used with dogs (Fig. 1). A 20-liter reservoir (1) containing 10% CO<sub>2</sub> in the resident gas was connected by the tube system to be studied (2) to an X-piece (3). This X-piece was the same used in dog experiments. It was connected on one side to a reciprocating pump (5), the frequency and amplitude of which could be varied. The second arm was connected to the tube system (2) leading to the reservoir. The third arm (4) was linked to the carrier gas source, e.g. a tank with either He, O<sub>2</sub>, or SF<sub>6</sub>. The fourth arm led to O<sub>2</sub>, CO<sub>2</sub> and/or N<sub>2</sub> meters (6) and through a needle valve (8) to a vacuum source. The CO<sub>2</sub> concentration in the X-piece was maintained at approximately 1% by flushing the X-piece with carrier gas (4). Thus, CO<sub>2</sub> was transported from the reservoir to the X-piece along a concentration gradient of approximately 10:1.

Frequency (f) was varied between 2 and 20 Hz. The average amplitude  $\Delta X$  of the oscillatory motion (tidal volume  $V_T$  divided by total cross-sectional area A) was varied between 15 and 50% of the length of the tube system used. Table 1 summarizes the geometry of the various systems used.

Amplitude and zero-line of the pressure in the reservoir was sensed with a Datametrics (7) pressure transducer and continuously monitored on an oscilloscope. The pressure readings served two purposes: (1) the amplitude was used to plethysmographically determine the tidal volume; it was calibrated with each gas mixture and at each frequency. (2) The average pressure in the reservoir was monitored and maintained throughout experiments at ambient pressure ( $\pm 0.01$  mm Hg) by adjusting the needle valve (8) controlling the outflow of gas. By maintaining ambient pressure in the reservoir at all times, bulk flow in the tube system was avoided.

Table 2 lists the gas properties of the gases used.  $D_m$  is the molecular diffusion coefficient or diffusivity of the gas being transported (O<sub>2</sub> or CO<sub>2</sub>) through a resident gas

(either He, N<sub>2</sub>, or SF<sub>6</sub>) with a kinematic viscosity  $\nu$ . The Schmidt number  $S$  is the ratio of  $\nu$  divided by  $D_m$ . Note that in this definition of  $S$ , the numerator and the denominator of  $S$  are not from the same gas. All measurements were made after 20 to 30 minutes of continuous running to allow for establishment of a quasi-steady state. In the quasi-steady state the concentration in the reservoir fell by about 0.02% CO<sub>2</sub>/min.

A compliant 2-liter bag (9) within a one-liter plexiglass cylinder was inserted between the X-piece and the reciprocating pump (Fig. 1) in order to seal the system from leaks through the pump. Before each experiment two leak tests were conducted, one based on a static pressure load, the second on running the system with pure O<sub>2</sub> and testing for the appearance of N<sub>2</sub> in the system. All measuring devices were calibrated before each experiment.

## RESULTS

We first report the data describing the transport of  $\text{CO}_2$  through either He,  $\text{O}_2$ , or  $\text{SF}_6$  in tubes of different radii. Fig. 2 shows data from tests done using capillaries with a radius of 0.047 cm, an oscillation amplitude of  $\Delta X = 5.47$  cm, and a frequency ranging from 2 to 20 Hz. Other details of the experimental conditions are given in Table 1. The conductance  $G$  in ml/min is plotted on the ordinate; it is defined as the amount of  $\text{CO}_2$  transported per unit of time and for a given concentration gradient ( $\dot{V}_{\text{CO}_2}/(F_{1\text{CO}_2} - F_{2\text{CO}_2})$ ).  $F_{1\text{CO}_2}$  is the fraction in the reservoir,  $F_{2\text{CO}_2}$  the fraction in the X-piece.  $G$  is analogous to alveolar ventilation and has the same units. It indicates how much bulk gas flow in ml/min would be needed to achieve the same  $\text{CO}_2$  elimination as obtained, presumably, by dispersion, in our system. Data obtained with He,  $\text{N}_2$ , or  $\text{SF}_6$  as resident gas show a curvilinear relationship between  $G_{\text{CO}_2}$  and  $f$ . Conductance of  $\text{CO}_2$  in  $\text{SF}_6$  is about 6 times higher than in He.

Fig. 3 shows data from tests done using a tube with a radius of 0.79 cm. The curvature of the three lines is less pronounced than in Fig. 2. Note that  $G_{\text{CO}_2}$  in He is now much higher than in  $\text{N}_2$  or  $\text{SF}_6$ . This observation is the reverse from the one in the capillaries. Note also that the conductance of  $\text{CO}_2$  in  $\text{SF}_6$  starts to decline when  $f$  exceeds 10 Hz.

For the next experiment (Fig. 4), the two pipe systems used in Figs. 2 and 3 are combined into a single system with the pipe with a radius of 0.79 cm being connected in series with the bundle of capillaries. The tidal volume is 30 ml, giving an oscillation amplitude of 11 cm in the capillaries and of 15.3 cm in the tube. The conductances of  $\text{CO}_2$  in He and  $\text{N}_2$  overlap; there is no clear separation of the two conductances, as seen in Figs. 2 and 3. The conductance in  $\text{SF}_6$  falls off the general pattern when  $f$  exceeds 10 Hz, as seen in Fig. 3.

A further experiment was conducted with beads. The external diameter of the beads was 0.244 cm, the calculated hydraulic radius 0.09 cm. The open channels between the beads therefore had an average radius slightly larger than the capillaries, but the geometry of these channels was irregular, including areas of narrowing, widening and branching. The beads were included in a 2-liter flask with a perforated bottom. The system was inserted between the X-piece and the reservoir shown in Fig. 1. The cross-section was  $67.3 \text{ cm}^2$ , and the estimated oscillation amplitude 0.45 cm. The results shown in Fig. 5 show a pattern quite similar to that found in the capillaries; the conductance in  $\text{SF}_6$  is about 2.9 times higher than in  $\text{N}_2$  and 6.4 times higher than in He.

## DISCUSSION

In 1983, Watson published a theoretical analysis of "diffusion in oscillatory pipe flow" (15), applying the concept of Taylor diffusion (14) to HFO in straight tubes. The theory is based on a number of assumptions such as laminar flow, incompressibility of the fluid, "infinite" tube length (i.e. no entrance effects), and very small concentration of the diffusing substance. Several papers have since experimentally confirmed various aspects of this study (4,5,6,9,10). The present amplifies these studies; this is the first study to use steady-state concentration gradients such as those that exist in the airways during steady-state gas exchange with gases at about the same concentrations as in biological systems. This is also the first study to analyze in detail the effect of gas properties in straight tubes of various sizes. The objective of this study was to obtain data that could be compared with similar tests done with animals. Some of the assumptions of Watson's theory are not met in the present experiments. The concentrations of the diffusing gases were much higher than in the theory. The flow was not always laminar; the conditions satisfied Epstein's criteria for laminarity in oscillatory flow (2), but the ratio of Reynold's number to  $\alpha$ , which is also suggested as a criterion for laminarity (13), was in some experiments above the critical value of 200. There were also, presumably, some undesired effects of compressibility since at high frequencies and high amplitudes the aerodynamic pressure drop across the studied tubes reached 1/10 atm. No correction was applied for any of the conditions that diverged from Watson's assumptions.

Watson (15) analyzes the gas transport in oscillatory flows in terms of two non-dimensional numbers, the Wormersley number ( $\alpha = a\sqrt{2\pi f/\nu}$ ) and the Schmidt number ( $S = \nu/D_m$ ). Both numbers interact in determining the gas transport and since each is made up of several factors, this implies that all these factors ( $a$ ,  $f$ ,  $\nu$ ,  $D_m$ ) interact. Thus, there is no simple rule that states the effect of resident gas viscosity on oscillatory flow. However,



there are two extreme conditions for which the effect of  $\nu$  is mathematically simple. These extreme conditions will be discussed first; the more general case will be presented later.

The theory predicts that the conductance is inversely proportional to the diffusivity of the diffusing gas if  $\alpha < 2$  (11). It also predicts that the conductance is proportional to the square root of the kinematic viscosity of the resident gas if  $\alpha > 20$ . Since the radius is the main factor determining  $\alpha$ , the above prediction applies in practice either to fairly small tubes or fairly large ones. The above theory may also be expressed in terms of the molecular weight of the resident gas; it implies that in small tubes the conductance increases with the square root of molecular weight of the resident gas, while in large tubes the conductance is inversely related to the square root of molecular weight. The inverse relationship for small and large tubes is surprising. It is due, in part, to large modifications of the velocity profile. In very small tubes and with  $\alpha < 2$ , the profile is rather similar to that in steady laminar flow. When  $\alpha$  is high, however, the profile is much more blunt and a boundary layer which is smaller than the radius becomes established. The value of  $\alpha$ , actually, is equal to the ratio of the radius divided by the boundary layer thickness.

For our experiments we chose conditions which are in accord with the above limits. In capillaries with  $a = 0.047$  cm, the value of  $\alpha$  is close to 2 (Table 1). The expected ratio of the conductance of  $\text{CO}_2$  in He and in  $\text{N}_2$  is equal to the inverse of the ratio of the respective molecular diffusion coefficients; according to Table 2, this ratio equals 0.25. The measured average ratio of the conductances in He and in  $\text{N}_2$  in Fig. 2 is  $0.26 \pm 0.02$ . Similarly, the expected ratio for the conductance of  $\text{CO}_2$  in He and in  $\text{SF}_6$  is 0.12; the measured average is  $0.17 \pm 0.02$ . Both sets of experiments are in agreement with the theory.

The predicted ratio of conductances in He and  $\text{N}_2$  using the tube with  $a = 0.79$  cm is 2.7 and our experimentally measured ratio is  $2.75 \pm .16$  (Fig. 3). The agreement with data obtained using  $\text{SF}_6$  is also good up to  $f = 10$  Hz. The conductance of  $\text{CO}_2$  in  $\text{SF}_6$ , however,

drops off above that frequency, an observation which is contrary to the theory. We believe the discrepancy to be due to turbulence. At  $f = 10$  Hz, the Reynolds number is 11,000 and the ratio  $Re/\alpha$  is 280. This is reason to suspect turbulence. Effects of compressibility may also have influenced our results.

The theory predicts a complicated and gradual change of the conductance in tubes in which  $\alpha$  is within the limits of 2 and 20 (10). This is illustrated in Fig. 6, which represents the normalized conductance  $\lambda$  plotted against  $\alpha$ . The conductance is normalized to permit comparisons between experiments done in tube systems of different lengths  $L$ , different total cross-sectional areas  $A$ , different frequencies  $f$  and different oscillation amplitudes  $\Delta X$  ( $\lambda = GL/A2\pi f\Delta X^2$ ). Two curves are shown, one for  $S = 1.86$  (diffusion of  $\text{CO}_2$  in He) and one for  $S = 1.0$  (diffusion of  $\text{CO}_2$  in  $\text{N}_2$ ). The value of  $\lambda$  is seen to rise with increasing  $\alpha$ , to reach a peak and then to fall again. This bell-shaped curve demonstrates how important  $\alpha$  is in determining  $\lambda$ . It implies a tuning effect that indicates that conductance is maximal at a given value of  $\alpha$  (10). The Schmidt number determines the location of the bell-shaped curve with respect to the abscissa. Since the two curves have a crossover, the ratio of  $\lambda$  for two gases is below unity on the left of the crossing over and above unity on the right of the crossing over.

Fig. 6 may be used to determine the effect of resident gas viscosity on transport for any tube size. Points A and B illustrate the condition at the lower end of the range of  $\alpha$ ; they depict the conditions in capillaries with  $a = 0.047$  cm,  $f = 20$  Hz, using He or  $\text{N}_2$ . The ratio of the conductances of  $\text{CO}_2$  in He and in  $\text{N}_2$  is 0.25, as mentioned earlier. This ratio, however, increases if the radius is increased. If the radius is increased, point A moves up along the  $S = 1.86$  curve, while point B moves up simultaneously on the  $S = 1.0$  curve. At some value of the radius, the ordinates become equal, i.e., there is no effect of resident gas density on dispersion (points C and D in Fig. 6). When the radius is further raised, the ratio

of conductances becomes larger than 1.0. It is noteworthy that the region for which the resident gas density is predicted to have no or only a minimal effect coincides with the region of the peak of the bell-shaped curve, i.e., the region of greatest dispersion.

Fig. 6 may be redrawn in terms of radius, which is the variable we are most interested in in the airways. Fig. 7 pictures the ratio of conductances  $G_{\text{in He}}/G_{\text{in N}_2}$  at  $f = 10$  Hz as a function of radius. The gradual change of the ratio from 0.25 to 2.7 is shown. For  $a = 0.25$  cm, the ratio is unity (this corresponds to points C and D in Fig. 6). The curve shown shifts to the left if frequency is raised or kinematic viscosity is lowered. Thus, the curve describing the ratio of  $G_{\text{in N}_2}/G_{\text{in SF}_6}$  is situated to the left of the curve shown with equality of conductances at  $a = 0.1$  cm. Experiments were conducted to confirm this in capillaries with  $a = 0.1$  cm. Identity of conductances in  $\text{N}_2$  and in  $\text{SF}_6$  was found at  $f = 14$  Hz rather than the predicted 10 Hz. These data are not shown for brevity.

How do these mostly theoretical considerations apply to the airways? We found in dog experiments that the change of resident gas from  $\text{N}_2$  to He and to  $\text{SF}_6$  has almost no effect on the gas exchange in HFOV when tidal volume and frequency are the same. These dog data are presented in a companion paper. In this paper we examine whether Taylor dispersion can explain these observations.

According to the theory, the effect of gas density varies strikingly with tube radius (Fig. 7). One may therefore wonder whether this effect is attenuated or whether it is even cancelled, if tubes of different sizes are connected in series. The theory would predict that the dispersion in tubes in series equals the sum of the dispersions in each tube ( $1/G_{\text{tot}} = 1/G_1 + 1/G_2$ ). Thus, attenuation is expected, if the conductances are matched and if transition effects at the connection site of the two tubes are negligible. We confirmed this in the experiment shown in Fig. 4. One may conjecture whether similar cancelling occurs in the

airways. Such a hypothesis assumes that the transport in short branching tubes such as the airways is similar to that in the long tubes used by the theory.

Paloski et al. have compared the transport in short branching tubes having an L/D ratio of 3.5 with that in long tubes (13). They find that Taylor dispersion plays an important role in gas transport, and that the transport in branched tubes is higher than predicted for straight tubes. They also find that the main features of the effect of resident gas density are the same in branching tubes and in straight ones: at small values of  $\alpha$ , the effect of resident gas is the reverse of the one at high values of  $\alpha$  with a gradual transition. If plotted in terms of Fig. 7, Paloski's data show a minimum ratio of 0.53 and a maximum ratio of 1.44. Thus, the pattern is the same though the magnitude of the effect of gas density is less. To that finding one may add our findings in beads that show that the dispersion in the multiple branching channels created between beads is quite similar to that of capillaries (Fig. 5). This supports the view that branching itself does not change the pattern of transport predicted by Taylor dispersion. The magnitude of  $\alpha$  is more important in determining the type of transport than the geometry of the channels. It is also noteworthy that cancellation of the effect of density occurs near a value of  $\alpha$  for which normalized conductance is maximal. In those airways in which density plays a role for the dispersion, the normalized dispersion is relatively low.

In conclusion, we believe that Taylor dispersion offers an explanation for our findings in dogs that replacing  $N_2$  by He or by  $SF_6$  has no significant effect on gas exchange. The dog data cannot be used to conclude that Taylor dispersion is not an important factor in gas transport.

We have emphasized in this paper the effect on dispersion of changing the resident gas. A closely related question has to do with the effect on dispersion of changing the diffusing gas. This question has been studied repeatedly in airways, perhaps most carefully by Knopp

et al. and Kaethner et al. (7,8). They found no difference in the washout of He, air, or SF<sub>6</sub> out of lungs during HFO and used these findings to conclude that the gas transport in HFO is not limited by a diffusion process.

Fig. 6 may be used to suggest another explanation for these experiments. The two curves shown define the normalized conductances of two different gases, if studied under the same conditions, i.e., in tubes of the same size, at the same frequency and with the same resident gas (i.e. with the same  $\alpha$ ). The curve labeled  $S = 1.86$  shows the conductance for a heavy gas such as SF<sub>6</sub> in air ( $S = \nu/D_m = 0.16/0.09$ ); the curve labeled  $S = 1.0$  is for a lighter gas such as CO<sub>2</sub> being dispersed in air ( $S = 0.16/0.16$ ). The two curves cross over. For values of  $\alpha$  below 3.5, the conductance of SF<sub>6</sub> exceeds that of CO<sub>2</sub>; for  $\alpha$  greater than 3.5, the conductance of CO<sub>2</sub> exceeds that of the heavier gas. It is somewhat surprising that a heavier gas can diffuse faster than a lighter one. The observation was already made by Taylor in his original paper for steady flow (14). We confirmed the finding in oscillatory flows (11,12).

One may now ask how two gases of different molecular weights are dispersed in a system of tubes of different sizes, such as the airways, for which the value of  $\alpha$  may vary below 3.5 in some parts of the system (tubes with a small diameter) to above 3.5 in some other part of the system. Since the conductances in the different parts of the system are additive, one may expect partial attenuation or even cancellation of the effect of the diffusivity of the diffusing gas. We believe therefore that the failure to detect differences in the washout of He, air, and SF<sub>6</sub> is in itself not a sufficient reason to conclude that diffusivity is not a limiting factor in the transport of gas in the airways. It may be a limiting factor in the transport in some part of the system and yet not be limiting for the airways as a whole.

**ACKNOWLEDGMENTS**

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TABLE 1

		a cm	A cm <sup>2</sup>	L cm	$\Delta X$ cm	f Hz	$\alpha$ for f=20 in N <sub>2</sub>
1)	capillaries	0.047	2.74	37	5.47	2-20	1.29
2)	capillaries	0.1	1.57	35	9.55	2-20	2.75
3)	tube	0.79	1.96	143	15.3	2-20	21.7
4)	beads, O.D. 0.244 cm	0.09	67.3	7	0.49	2-20	2.6

TABLE 2: GAS PROPERTIES

	He	N <sub>2</sub>	SF <sub>6</sub>
<b>Experiments in which resident gas is varied.</b>			
Kinematic viscosity $\nu$ (cm <sup>2</sup> /sec)	1.17	.167	.024
Diffusion coefficient $D_m$ (cm <sup>2</sup> /sec) of CO <sub>2</sub> in*	.63	.16	.074
Schmidt number $\nu/D_m$ for CO <sub>2</sub> in	1.86	1.00	.32

\* From Bird, Stewart, and Lightfoot, Transport Phenomena, New York, Wiley, 1960, p. 505.

## LEGENDS TO FIGURES

- Figure 1: Schematic of experimental setup. 1) Container with 10% CO<sub>2</sub> in either He, N<sub>2</sub> or SF<sub>6</sub>. 2) Tube system. 3) X-piece where a concentration of CO<sub>2</sub> of about 1% is maintained. CO<sub>2</sub> is transported by augmented diffusion from the container to the X-piece. 4) Inflow of resident gas. 5) Reciprocating pump. 6) Outflow of gas with vacuum source, throttle (#8) and gas-measuring devices. 7) Pressure transducer measuring pressure in container is used to determine tidal volume.
- Figure 2: Conductance of CO<sub>2</sub> in capillary tubes with  $a = 0.047$  cm in three resident gases (He, N<sub>2</sub>, and SF<sub>6</sub>). Oscillation amplitude 5.47 cm; tube length 37 cm. The conductance  $G$  is defined by  $G = \dot{V}_{CO_2}/(F_1 - F_2)$ .
- Figure 3: Conductance of CO<sub>2</sub> in a single tube of 143 cm in length and .79 cm ID with three different resident gases. Oscillation amplitude: 15.3 cm.
- Figure 4: Conductance of CO<sub>2</sub> in a system made up of the single tube and the capillaries connected in series.
- Figure 5: Conductance of CO<sub>2</sub> in a system made up of beads with O.D. = 0.244 cm. Length of the bed of beads: 7 cm; oscillation amplitude: 0.49 cm.
- Figure 6: Relationship between the normalized conductance  $\lambda$  and  $\alpha$  derived (8) from Watson's theory (13). The relationships for  $S = 1.86$  (diffusion for CO<sub>2</sub> in He) and for  $S = 1.0$  (diffusion of CO<sub>2</sub> in N<sub>2</sub>) are shown.

Figure 7: Relationship between the ratio of the conductances of  $\text{CO}_2$  in He and in  $\text{N}_2$  ( $G_{\text{in He}}/G_{\text{in N}_2}$ ) and tube radius.

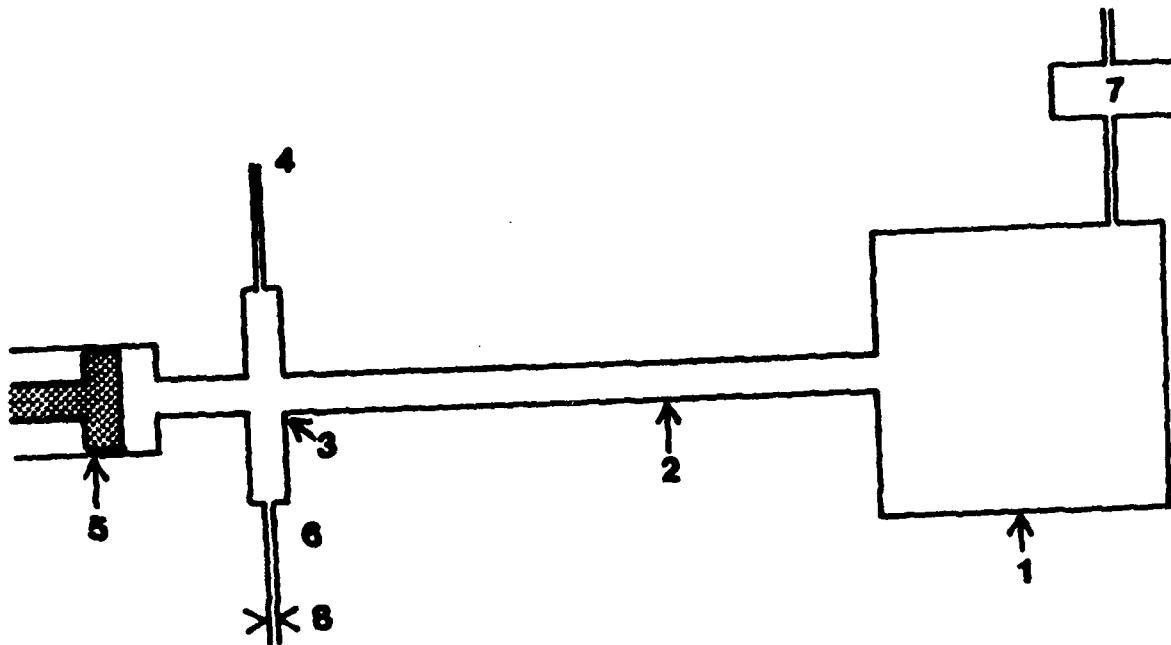


Fig. 1

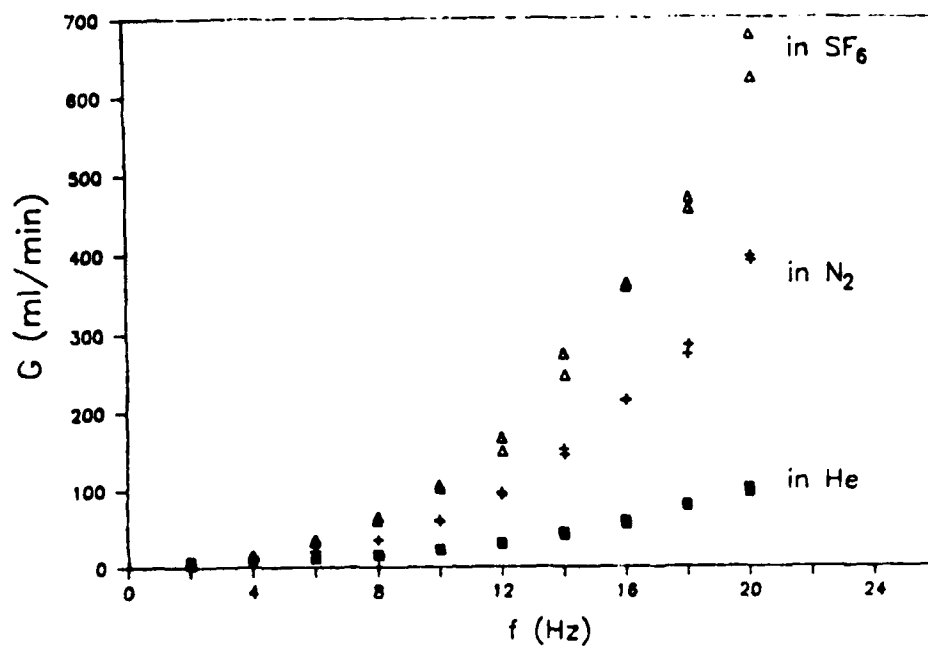


Fig. 2

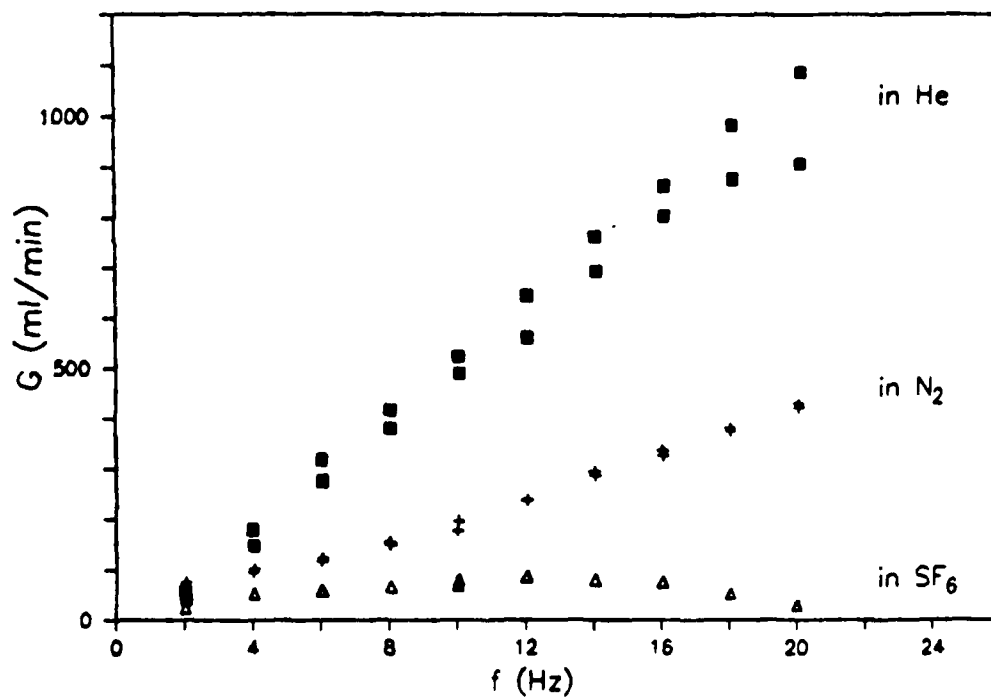


Fig. 3

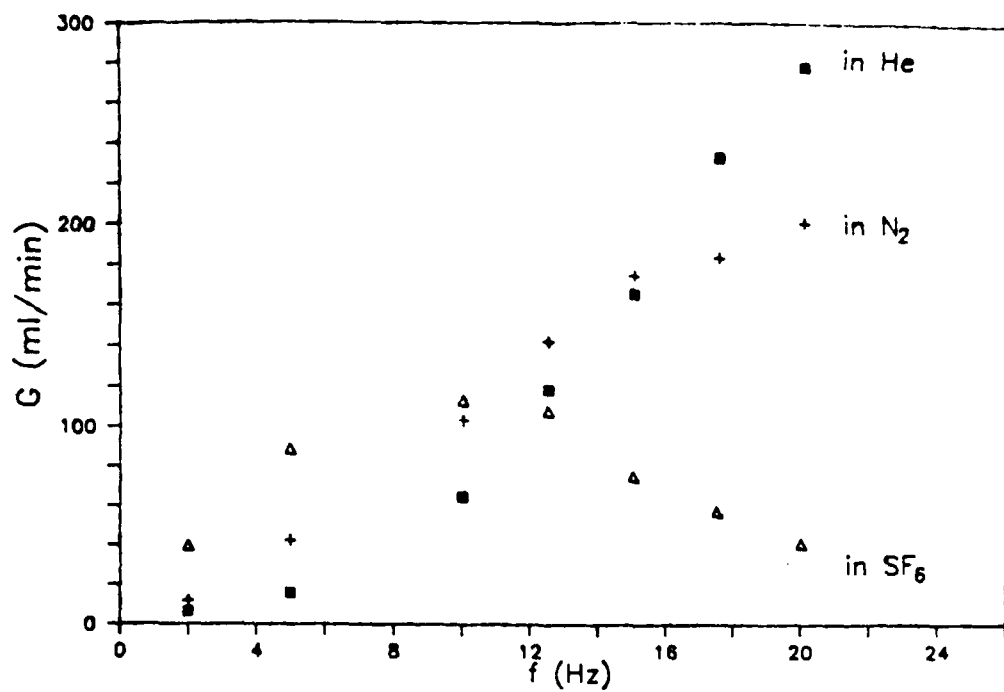


Fig. 4

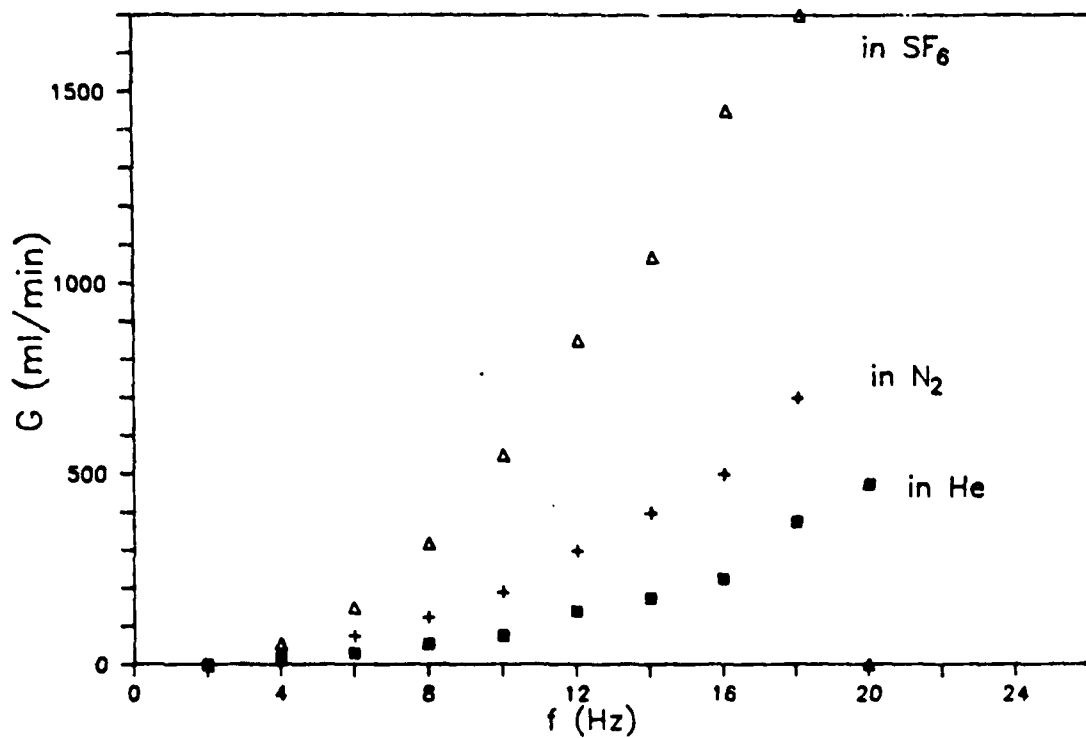


Fig. 5

A

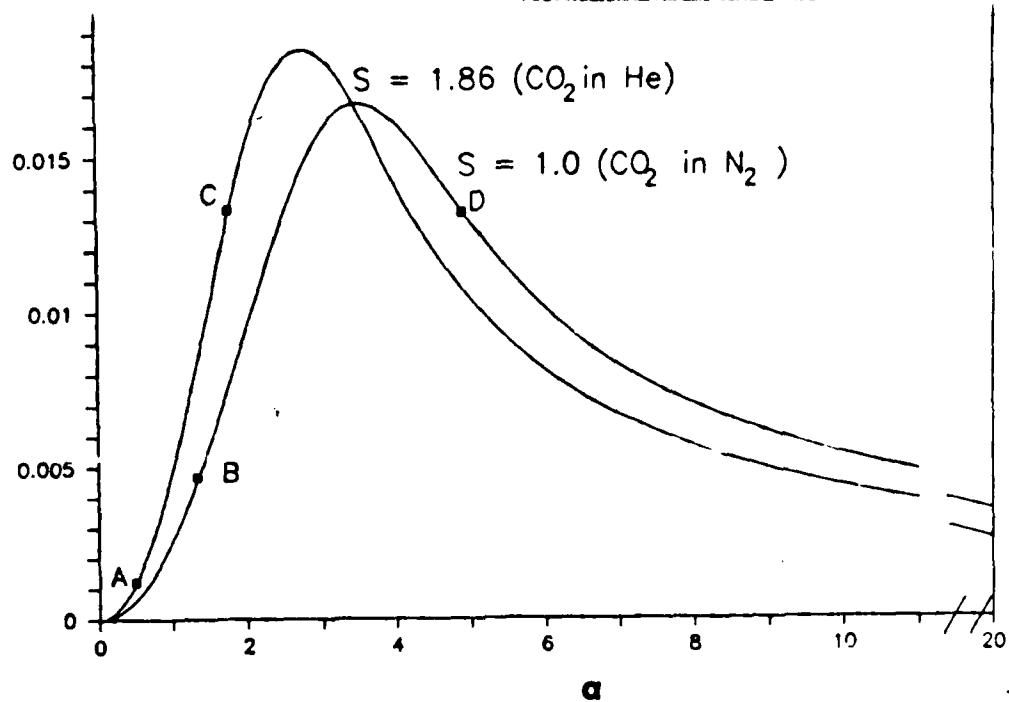


Fig 6

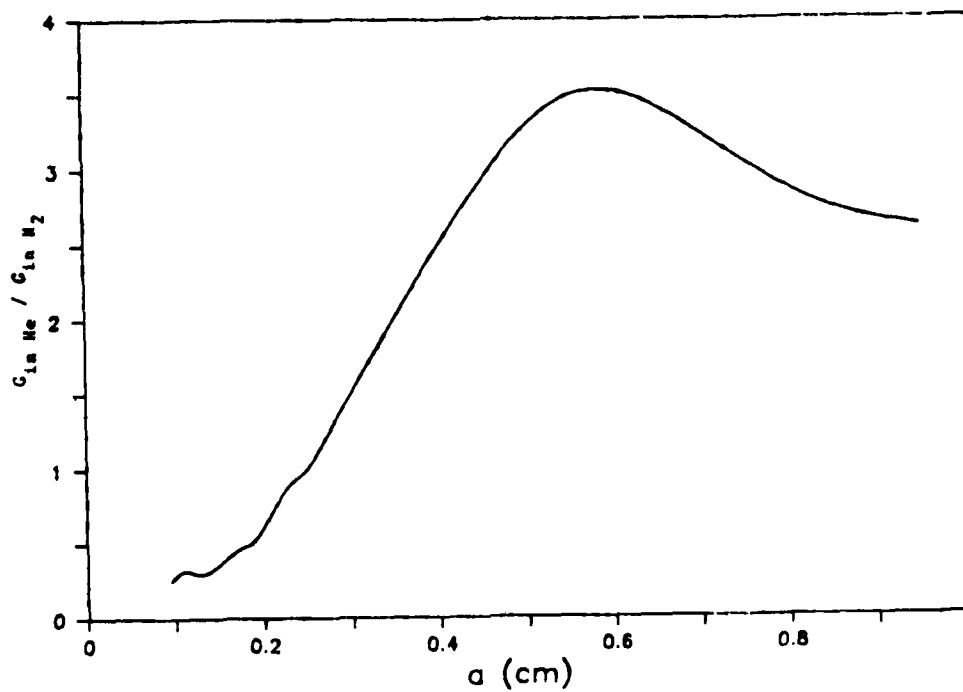


Fig. 7



DIFFUSIONAL SEPARATION OF GASES AND SOLUTES  
IN OSCILLATORY FLOW

by Marc J. Jaeger, Thomas Soepardi, and Ali Maddahian

Department of Physiology  
University of Florida

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## INTRODUCTION

Taylor (14) has shown that the spreading of a substance introduced in small quantity into a fluid flowing in a pipe is greatly enhanced by the fluid velocity. This enhancement is the result of the combined action of radial molecular diffusion and the variation of velocity over the cross section. It is especially high in oscillating, laminar flow because of the particular time-dependent velocity profile in this type of flow. Oscillatory flow has the further advantage that there is no net, continuous flow in the pipe and that the spreading occurs, if averaged over time, from a stationary site. The increased spreading can be expressed as effective diffusion or dispersion (3-7,16). The effective diffusion coefficient is 4 to 5 orders of magnitude greater than the molecular diffusion coefficient of the same substance (6). The process can be used to separate substances with different molecular diffusion coefficients (8). In this study we present data obtained for gases and liquids. Some early work by Dreyer et al. (1) will be discussed below.

## THEORY

Watson (16) showed that the effective diffusion coefficient,  $D_{eff}$ , in oscillatory pipe flow is a function of (a) the molecular diffusion coefficient  $D_m$  of the tracer in the fluid, (b) the kinematic viscosity,  $\nu$ , of the carrier fluid, (c) the radius,  $a$ , of the cylindrical pipe, and (d) the angular velocity,  $\omega$ , of the oscillations. For convenience the above parameters are combined into nondimensional numbers: the Schmidt number  $\sigma = \nu/D_m$ , and the Wormersley number  $\alpha = a\sqrt{\omega/\nu}$ . The transport equation for a straight, cylindrical pipe with length  $L$  and cross-sectional area  $A$  may be written, in analogy to the conventional diffusion equation, as (3-5):

$$G = \dot{q} / (c_1 - c_2) = D_{eff} A / L \quad \text{Eq. 1}$$

where  $\dot{q}$  is the rate of diffusional transport,  $c_1$  and  $c_2$  are the concentrations of the diffusing substance at each end of the diffusing path, and  $D_{eff}$  is the effective diffusion coefficient. Watson found that  $D_{eff}$  is proportional to the square of the oscillation amplitude  $\Delta X$  ( $\Delta X =$

stroke volume/cross-sectional area). For convenience,  $D_{eff}$  is normalized by the product of the oscillation amplitude squared and angular frequency,  $\lambda = D_{eff}/\omega\Delta X^2$ .

The normalized diffusion coefficient  $\lambda$  is, under certain limiting conditions, a simple function of  $\alpha$  and  $\sigma$ . These limiting conditions are different for gases and liquids and are tabulated for clarity in Table 1:

TABLE 1.

Gas	$\alpha < 2$	$\alpha > 20$
$0.1 < \sigma < 2.0$	$\lambda = \sigma\alpha^2/384$	$\lambda \approx \frac{1}{12\sqrt{2}\alpha}$
Liquid	$\alpha < 0.1$	$\alpha > 1$
$500 < \sigma < 5000$	$\lambda = \sigma\alpha^2/384$	$\lambda \approx \frac{1}{4\sqrt{2}\sigma\alpha}$

If the limiting conditions do not apply, the more general formulation must be used (6,11):

$$\lambda(\alpha, \sigma) = D_{eff}/\omega\Delta X^2 = AB/C \quad \text{Eq. 2}$$

with  $A = \sigma/4\alpha(\sigma^2 - 1)$

$$B = F_i(\alpha) - \frac{1}{\sqrt{\sigma}} \left[ \frac{|F(\alpha)|}{|F(\alpha\sqrt{\sigma})|} \right]^2 F_i(\alpha\sqrt{\sigma}); \quad F(\alpha) = F_R(\alpha) + iF_i(\alpha)$$

$$C = |1 + 2F(\alpha)/\alpha|^2$$

$$F_R(\alpha) = [\text{ber}\alpha \text{ber}'\alpha - \text{ber}\alpha \text{bei}'\alpha]/[\text{ber}^2\alpha + \text{bei}^2\alpha]$$

$$F_i(\alpha) = [\text{ber}\alpha \text{ber}'\alpha + \text{bei}\alpha \text{bei}'\alpha]/[\text{ber}^2\alpha + \text{bei}^2\alpha]$$

Note that the theory predicts that  $\lambda$  (and therefore  $D_{eff}$ ) may be either proportional to  $D_m$  (in liquids if  $\alpha > 1$ ) or inversely proportional to  $D_m$  (in liquids and in gas if  $\alpha$  is small; 8).

Dispersion is quasi-independent of  $D_m$  in gases if  $\alpha > 20$ ; thus gas separation is not feasible for  $\alpha > 20$ .

Separation of gases in a gaseous carrier or of solutes in a liquid carrier can be achieved whenever the ratio of the normalized diffusion coefficients for the two substances is not unity. Optimal separation, however, exists only under narrowly defined conditions. To illustrate these optimal conditions, we solve Eq. 2 for gases (Figs. 1 and 2) and for liquids (Figs. 3 and 4). In Fig. 1 we assume two gases labeled H and L, for heavy and light, which are to be separated by augmented diffusion in a third gas, the resident gas ( $O_2$ ), with a kinematic viscosity of  $\nu = 0.16 \text{ cm}^2/\text{sec}$ . The light gas is helium,  $\sigma_L = 0.2$ , and the heavy gas is  $CO_2$ ,  $\sigma_H = 1.0$ . The normalized dispersion coefficients,  $\lambda_H$  and  $\lambda_L$ , are plotted for various values of  $\alpha$  in Fig. 1. The maximal values for  $\lambda_H$  and  $\lambda_L$  are at  $\alpha = 4.0$  and  $6.0$ , respectively. The relative magnitudes of  $\lambda_H$  and  $\lambda_L$  are illustrated in Fig. 2, in which  $\lambda_L/\lambda_H$  and  $(\lambda_H - \lambda_L)$  are plotted against  $\alpha$ . It is noted that  $\lambda_L/\lambda_H$  approaches a minimum for  $\alpha < 1$ ; this minimum is equal to  $D_{mH}/D_{mL} = 0.20$  (Table 1). The difference,  $\lambda_H - \lambda_L$ , has a maximum at  $\alpha = 3.1$  and declines rapidly at values of  $\alpha$  above and below  $3.1$ . At  $\alpha = 3.1$  the ratio  $\lambda_L/\lambda_H$  is only slightly higher ( $0.27$ ) than its lowest value for  $\alpha < 1$ . Selection of the best operating point involves two considerations: optimal separation ( $\lambda_L/\lambda_H$ ) and optimal differential diffusion ( $\lambda_H - \lambda_L$ ). According to Fig. 2, this value is approximately  $\alpha = 3.1$ . The peak for  $(\lambda_H - \lambda_L)$  defines the value of  $\alpha$  for which the differential flux  $\dot{q}_H - \dot{q}_L$  is likely to be highest. Note that this value of  $\alpha$  can be obtained with various combinations of  $a$ ,  $\omega$ , and  $\nu$ ; this permits optimization and free choice of the carrier fluid.

The separation of solutes in water is illustrated in a similar manner by Figs. 3 and 4. We chose two solutes ( $\sigma_H = 2000$  and  $\sigma_L = 500$ ) diffusing in water ( $\nu = 0.01 \text{ cm}^2/\text{sec}$ ). Comparison of the normalized diffusion coefficient of gases and solutes during HFO reveals the following:

1. The maximal values reached by  $\lambda$  for gaseous and for liquid dispersion are nearly the same: for  $\sigma = 0.3$ ,  $\lambda_{\max} = 0.00790$ ; for  $\sigma = 2000$ ,  $\lambda_{\max} = 0.019$ .
2. Maximal dispersion is reached in liquids at much lower values of  $\alpha$  than in gases. Maximal dispersion in water as compared to gas is reached if  $\alpha_{\text{H}_2\text{O}}/\alpha_{\text{gas}} = \sqrt{\sigma_{\text{H}_2\text{O}}/\sigma_{\text{gas}}}$ .
3.  $\lambda_L/\lambda_H$  varies in liquids between a minimum equal to  $D_{mH}/D_{mL}$  and a maximum equal to  $D_{mL}/D_{mH}$ . In gases, the maximal value is approximately unity.
4. The difference  $(\lambda_H - \lambda_L)$  has identical positive and negative maxima in liquids; in gases, the negative maximum is negligibly small.

From Fig. 4 one would predict best separation of the chosen solutes H and L at  $\alpha = 0.07$  for which  $(\lambda_H - \lambda_L)$  has a maximum while  $\lambda_L/\lambda_H$  equals 0.35, close to its minimum of 0.2. At  $\alpha = 0.22$ ,  $(\lambda_H - \lambda_L)$  has its negative maximum. Note that maximal and minimal values of  $(\lambda_H - \lambda_L)$  occur within a narrow range of values of  $\alpha$  ( $0.07 \leq \alpha \leq 0.22$ ).

## METHODS

The experimental setups for gaseous and liquid separation are similar (Fig. 5). They consist of a reciprocating pump that generates oscillatory flows in capillary tubes. A reservoir at one end of the capillaries holds the fluid (gaseous or liquid) which contains the two substances that are to be separated. The area at the other end of the capillaries is flushed with pure carrier fluid at a rate  $Q$ . This generates a concentration gradient for the diffusing substances H and L, which are transported at rates  $\dot{q}_H$  and  $\dot{q}_L$  from the container toward the area being flushed. The partially separated substances are collected and analyzed at the outflow of the system. The continuity equation requires the carrier fluid to diffuse in the opposite direction at a rate  $\dot{q}_c = \dot{q}_H + \dot{q}_L$ .

The specific conditions for gaseous separation were as follows: We chose a bundle of 76 capillaries with radius  $a = 0.19$  cm and length  $L = 120$  cm. The value of  $\alpha$  was varied between 2.66 and 4.61. The oscillation amplitude was less than 20% of  $L$ . We used a gas mixture of 10% He, 10% CO<sub>2</sub> and 80% O<sub>2</sub>, O<sub>2</sub> being the carrier gas. The reservoir had a volume of 20 L. The pressure in the reservoir was continuously monitored and kept at an average value of  $P_{amb} \pm 0.01$  mm Hg by adjusting the outflow to match  $Q$  exactly.

The system for liquid separation consisted of a bundle of 91 capillaries with  $a = 0.087$  cm and  $L = 120$  cm. Alpha was varied between 0.52 and 0.89. The oscillation amplitude was kept below 40% of  $L$ . The reservoir had a volume of 0.2 L and had a distensible top. We used the following solutes: KCl, CuSO<sub>4</sub>, glucose, Methylene Blue.

Experimental separation depends on  $Q$ , the rate of flushing with carrier fluid. If  $Q$  is high, the available concentration gradient is high and the dispersion is maximal. This provides for best separation, but the concentration of the diffusing substances in the outflow is low. On the contrary, if  $Q$  is reduced, the dispersion and separation are reduced as well. It may be shown (8) that the separation factor  $\rho_L$  equals:

$$\rho_L = C_{L2}C_{H1}/C_{L1}C_{H2} = (G_L/Q + \lambda_L/\lambda_H)/(G_L/Q + 1) \quad \text{Eq. 3}$$

We chose to vary  $G_L/Q$  between 0.05 and 0.5.

## RESULTS AND DISCUSSION

### 1) Separation of gases:

Table 2 shows gas separation data obtained with He and CO<sub>2</sub> diffusing through the carrier, O<sub>2</sub>. The Wormersley number  $\alpha$  was chosen close to the optimal value of 3.5 (Fig. 2). A good agreement between experimental and predicted data is found. A high value of  $Q$  was used to get the best possible separation (Eq. 3). Fig. 6 shows the effect of  $Q$  on  $G$ , as predicted by Eq. 3. In Fig. 7,  $(G_H - G_L)$  is found to be proportional to  $V_T^{1.87}$ , a value slightly lower than the expected square relationship (Eq. 2). Maximal dispersion  $D_{eff} = G_H L/A$  was 1032 cm<sup>2</sup>/sec or 6470 times the molecular diffusion coefficient of CO<sub>2</sub> in O<sub>2</sub>. The tidal volume  $V_T$  was determined from the pressure variation in the container.

The experimental conditions used for these experiments differ appreciably from the theory used to analyze them. Watson assumes incompressible flow. Compressibility effects are notable in the system used, in which the pressure drop in the capillaries is up to 0.1 atm. Moreover, Watson assumes only trace amounts of the diffusing substance, thus maintaining the kinematic viscosity constant along the diffusing path. Fairly large concentrations of He and CO<sub>2</sub> are used in our system, a fact that results in a continuous change of the kinematic viscosity along the diffusing path. Moreover, the theory does not account for entrance effects, which, presumably, determine the velocity profile for some length at each end of the capillaries. Entrance effects may reduce separation.

## 2) Separation of solutes:

Separation of solutes in oscillatory liquid flow was achieved by Dreyer et al. long before the recent interest in this field (1). Dreyer et al. developed an equation similar to the one in Table 1 for  $\alpha < 0.1$ , based on the simplified assumption that the velocity profile is paraboloid. They found good separation using a single capillary with  $a = 0.025$  cm and  $L = 18$  cm at  $\alpha = 0.61$ . Their dispersion coefficient was nearly 11,000 times higher than  $D_m$ . Kurzweg recently summarized the theory (10); his suggestions for optimal separation are different from ours. We confirmed with a large number of experiments that  $D_{eff}$  is proportional to  $\Delta X^2$ .  $D_{eff}$  for KCl was  $17.4 \text{ cm}^2/\text{sec}$  or 870,000 times  $D_m$  of KCl in  $\text{H}_2\text{O}$ . The predicted  $D_{eff}$  under the given conditions was  $\lambda\omega\Delta X^2 = 0.00212 \times 1.04 \times (61.4)^2 = 8.31 \text{ cm}^2/\text{sec}$  (Eq. 2). Results obtained with KCl ( $\sigma = 500$ ),  $\text{CuSO}_4$  ( $\sigma = 1136$ ), glucose ( $\sigma = 1450$ ) and Methylene Blue ( $\sigma = 1923$ ) are shown in Fig. 7. The  $\dot{q}_{\text{KCl}}$  reached a maximum of 29 mM/min when [KCl] was 0.075 N.

## CONCLUSION

The evidence presented indicates that gases and solutes may be separated in oscillatory fluid flow. The separation is close to the ratio of the molecular diffusion coefficients. The enhancement of the flux in our experiments was 3 orders of magnitude in the gaseous phase and close to 6 orders of magnitude in the liquid phase. The energy consumption was low; in the gaseous experiments it was 14 Watts. The method allows free choice of carrier fluid. Best choice of carrier fluid is, presumably, one that can be removed easily and cheaply (water vapor,  $\text{CO}_2$ ).

The process may be appreciably enhanced over and above the optimization already achieved. The stroke length  $\Delta X$  can be increased beyond the values used until turbulence occurs. No separation is expected in turbulent flow (15). The criteria for turbulence in



oscillatory flow are not yet as well defined as in steady flow (12,13). From the data provided and from newer information (11), it would seem that  $\Delta X$  can be raised appreciably over the values used, permitting an increase of the dispersion of perhaps 1 order of magnitude. Moreover, the concentrations of the gases to be separated may be raised. In a preliminary experiment we raised them to 50/50, eliminating the need for a carrier fluid in the container. The  $\dot{q}_H$  was raised from 44 to 588 ml/min by this modification without a decrease of the separation factor  $\rho_L$ .

The process shown is suitable for continuous, steady-state operation, if the fluid in container 1 is continuously replenished. The experiments quoted in which no carrier gas was added to container 1 were run in this fashion.

The study of substance separation by dispersion provides interesting insight into the mechanism of dispersion. It may be seen from Figs. 1 and 3 and prior work (7) that maximal dispersion occurs when  $\alpha^2\sigma = \pi^2$ . This relationship illustrates the interplay of diffusion and fluid mechanics which is characteristic for this phenomenon and may be looked at from two viewpoints: 1)  $\alpha^2\sigma = \pi^2$  is identical to  $\delta/a = \sqrt{\sigma}/\pi$ , where  $\delta$  is the Stokes boundary layer thickness; thus, optimal dispersion occurs when the thickness of the boundary layer relative to the radius has a fixed relationship to  $\sigma$ . The Schmidt number  $\sigma$  is close to unity in gases, resulting in  $\delta/a$  equalling about  $1/\pi$ . In liquids, however,  $\delta/a$  is approximately equal to  $\sqrt{1000}/\pi$ . 2) The same relationship may also be written as  $t_d/t_e = \pi$  where  $t_d$  is the time available for radial diffusion ( $a^2/D_m$ ) and  $t_e$  is the time of a half cycle.

TABLE 2.  
Gas Separation Data

$\alpha$	$\rho_L^*$ (experimental)	$\rho_L^*$ (theory)	$G_L/Q$
2.66	$0.40 \pm 0.03$ (N=8)	0.28	0.06
3.76	$0.43 \pm 0.08$ (N=6)	0.44	0.13
4.61	$0.49 \pm 0.15$ (N=5)	0.56	0.13

\* see Equation 3.

### ACKNOWLEDGMENTS

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### LEGENDS TO THE FIGURES

- Fig. 1: The normalized dispersion coefficient  $\lambda = D_{\text{eff}}/\omega\Delta X^2$  is plotted against the Wormersley coefficient  $\alpha = a\sqrt{\omega/\nu}$  for CO<sub>2</sub> diffusing in O<sub>2</sub> ( $\sigma = 1.0$ ) and He diffusing in O<sub>2</sub> ( $\sigma = 0.2$ ).
- Fig. 2:  $\lambda_{\text{He}}/\lambda_{\text{CO}_2}$  and  $\lambda_{\text{CO}_2} - \lambda_{\text{He}}$  are plotted against  $\alpha$ .
- Fig. 3: The normalized dispersion coefficient  $\lambda$  is plotted against  $\alpha$  for two solutes in water, one heavy with  $\sigma_{\text{H}} = 2000$  and one light,  $\sigma_{\text{L}} = 500$ .
- Fig. 4:  $\lambda_{\text{L}}/\lambda_{\text{H}}$  and  $\lambda_{\text{H}} - \lambda_{\text{L}}$  are plotted against  $\alpha$  for the same solutes used in Fig. 3.
- Fig. 5: Experimental setup showing a reservoir with the gases to be separated, the capillaries which serve as diffusing path, the inflow of carrier fluid (Q) and the pump.
- Fig. 6: Effect of dilution by the carrier fluid flow Q on the separation. Shown as a line is the theoretical relationship. Circles are experimental values of  $G_{\text{He}}/G_{\text{CO}_2}$ .
- Fig. 7: Effect of tidal volume  $V_{\text{T}}$  (measured indirectly as pressure changes in the reservoir) on the differential separation  $G_{\text{CO}_2} - G_{\text{He}}$ .
- Fig. 8: Separation of 5 pairs of solutes. The numbers indicate the number of experiments.

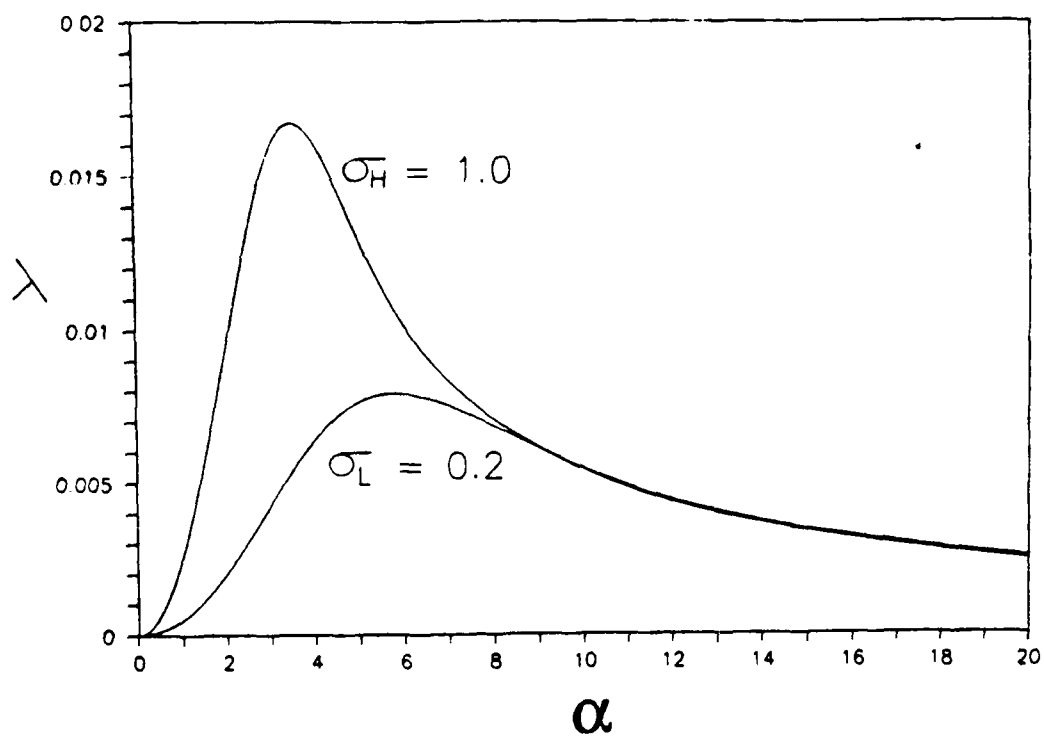


Fig. 1

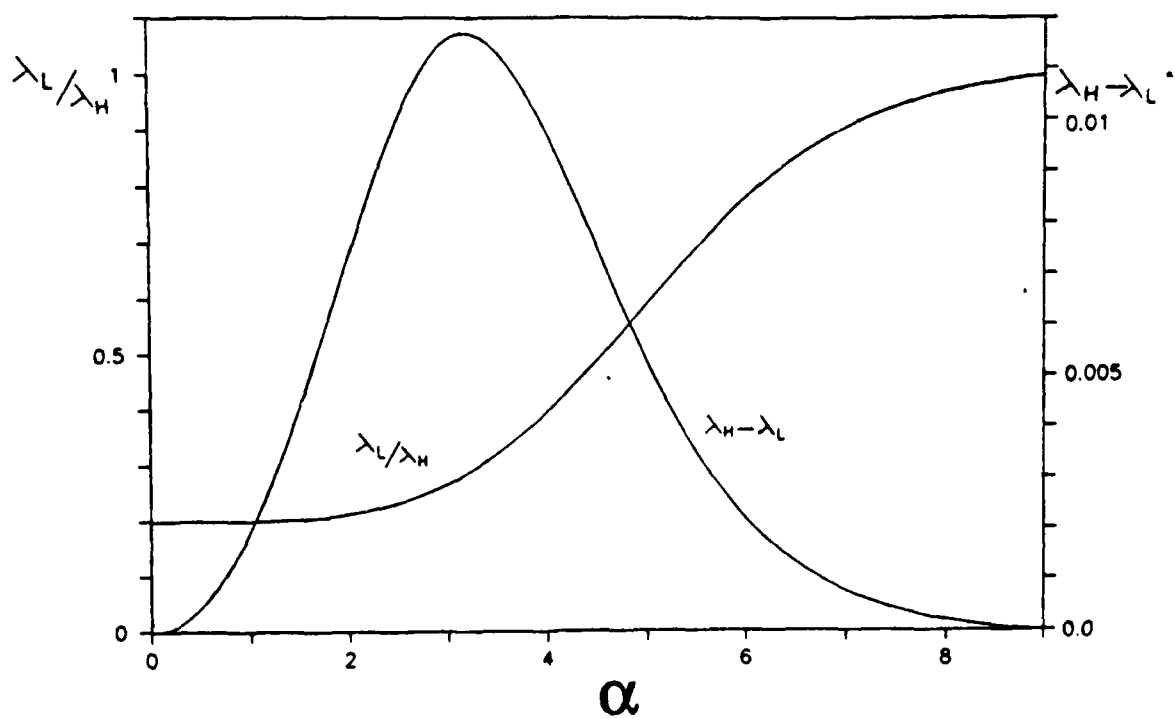


Fig. 2

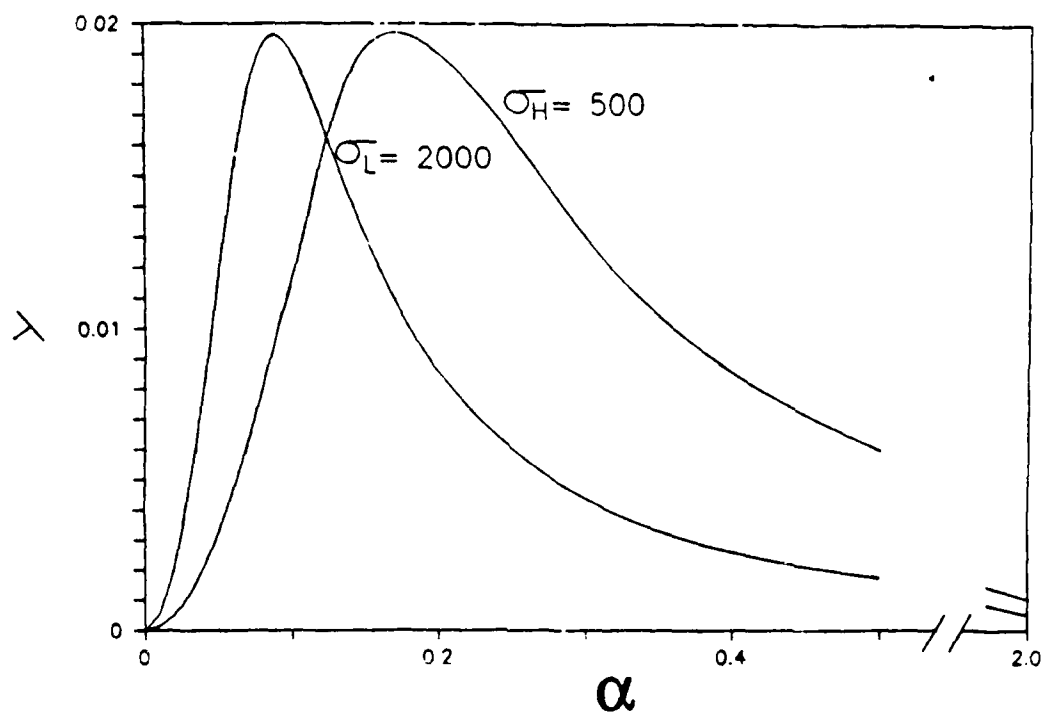


Fig. 3

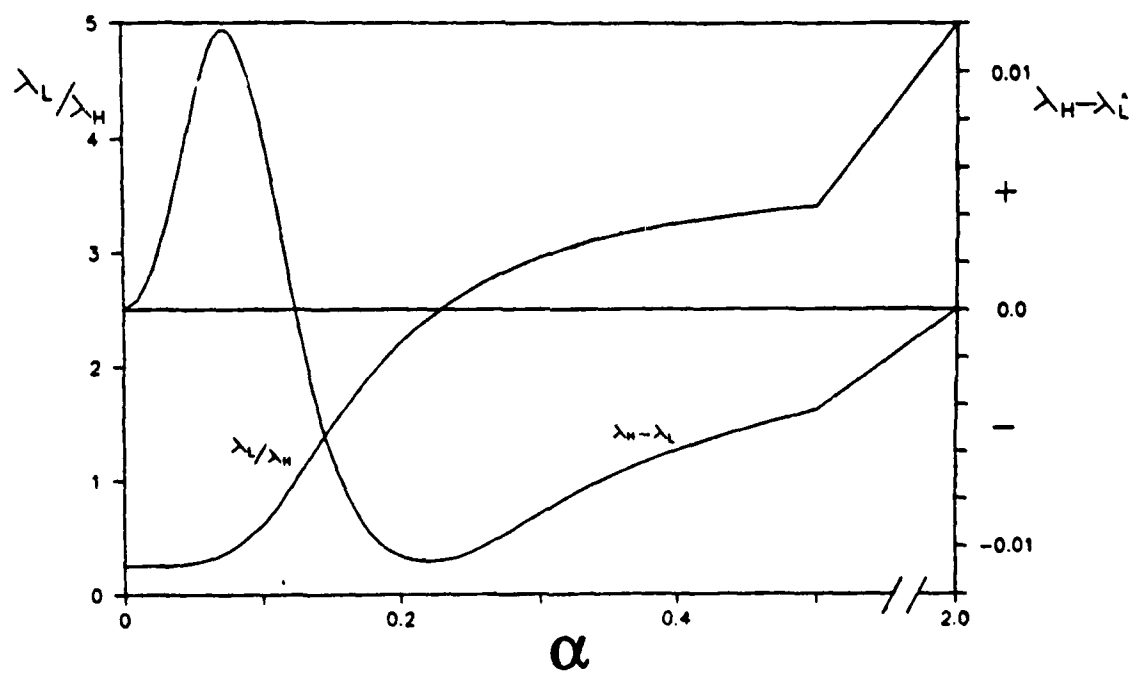


Fig. 4



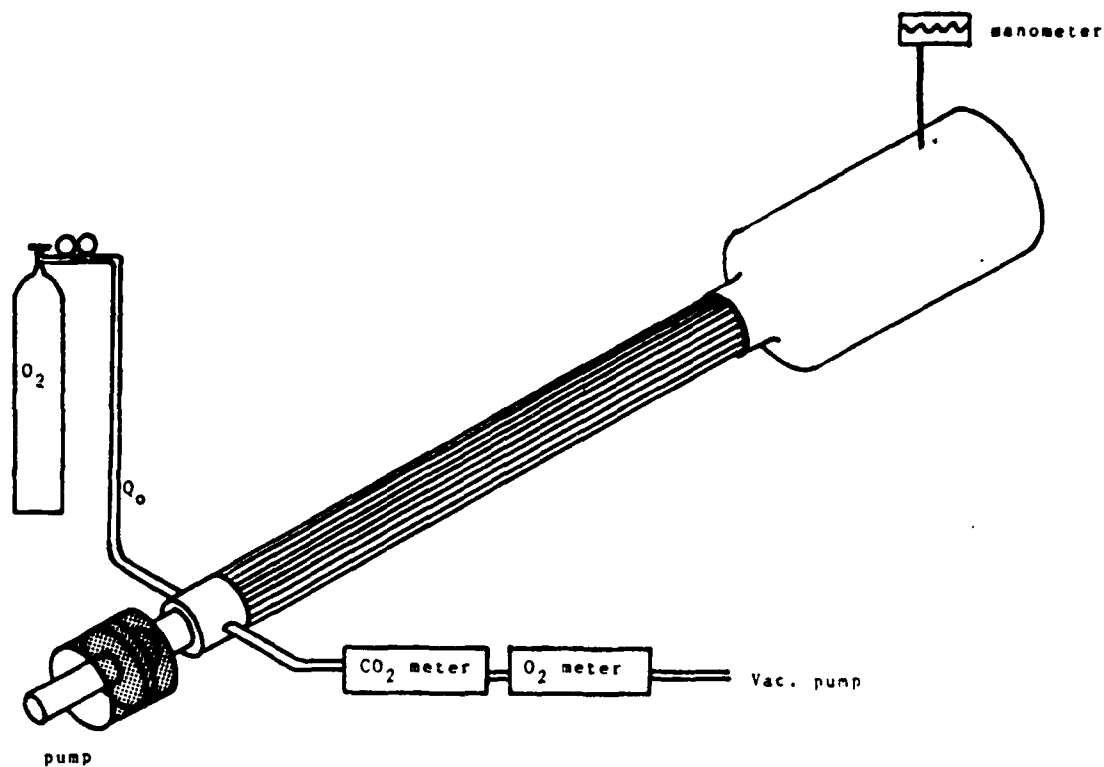


Fig. 5

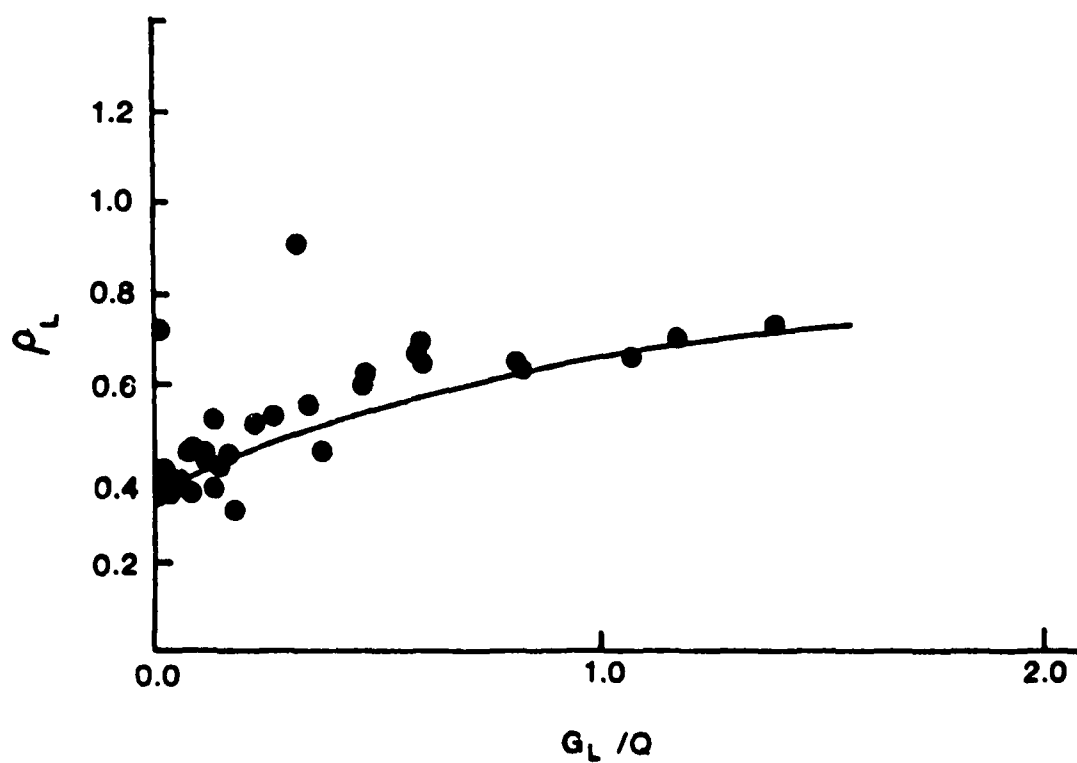


Fig. 6

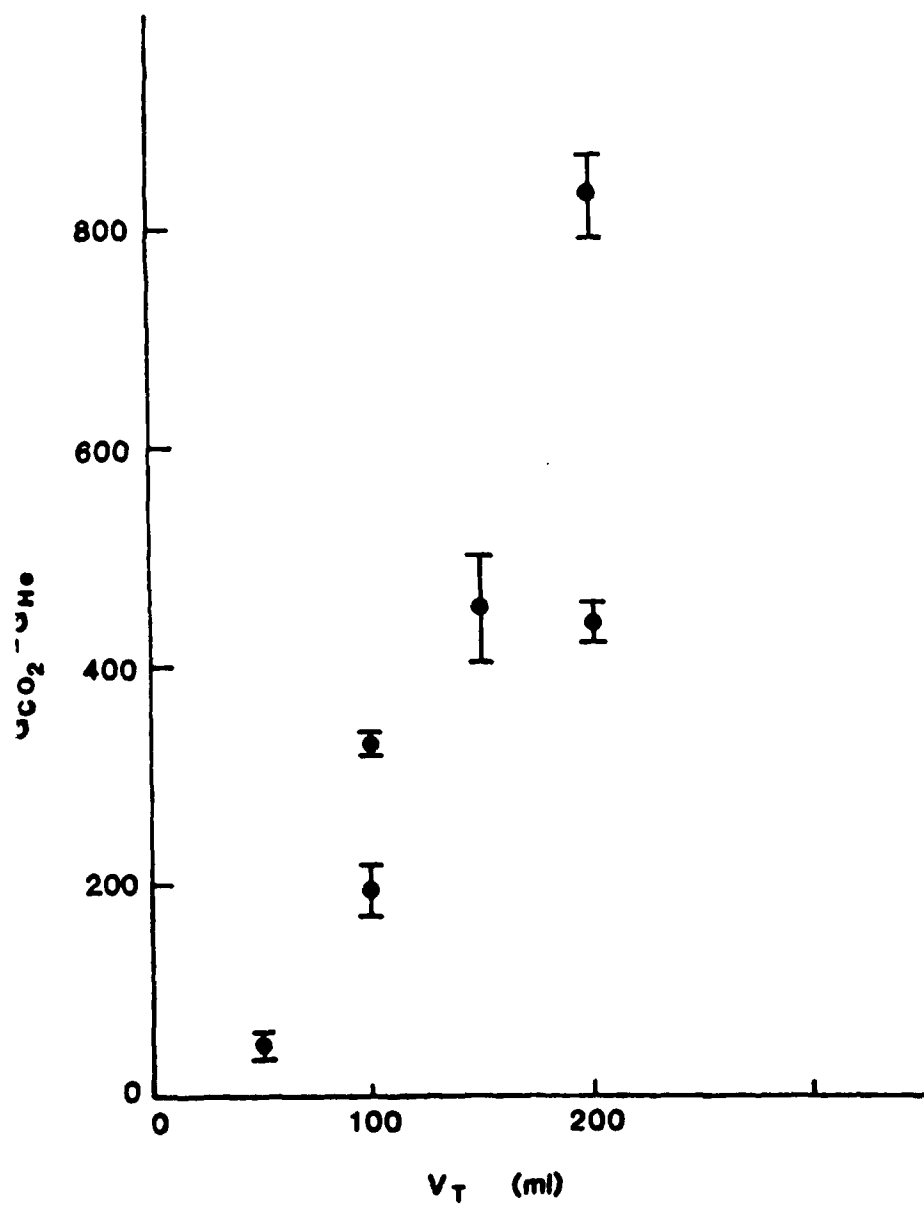


fig. 7

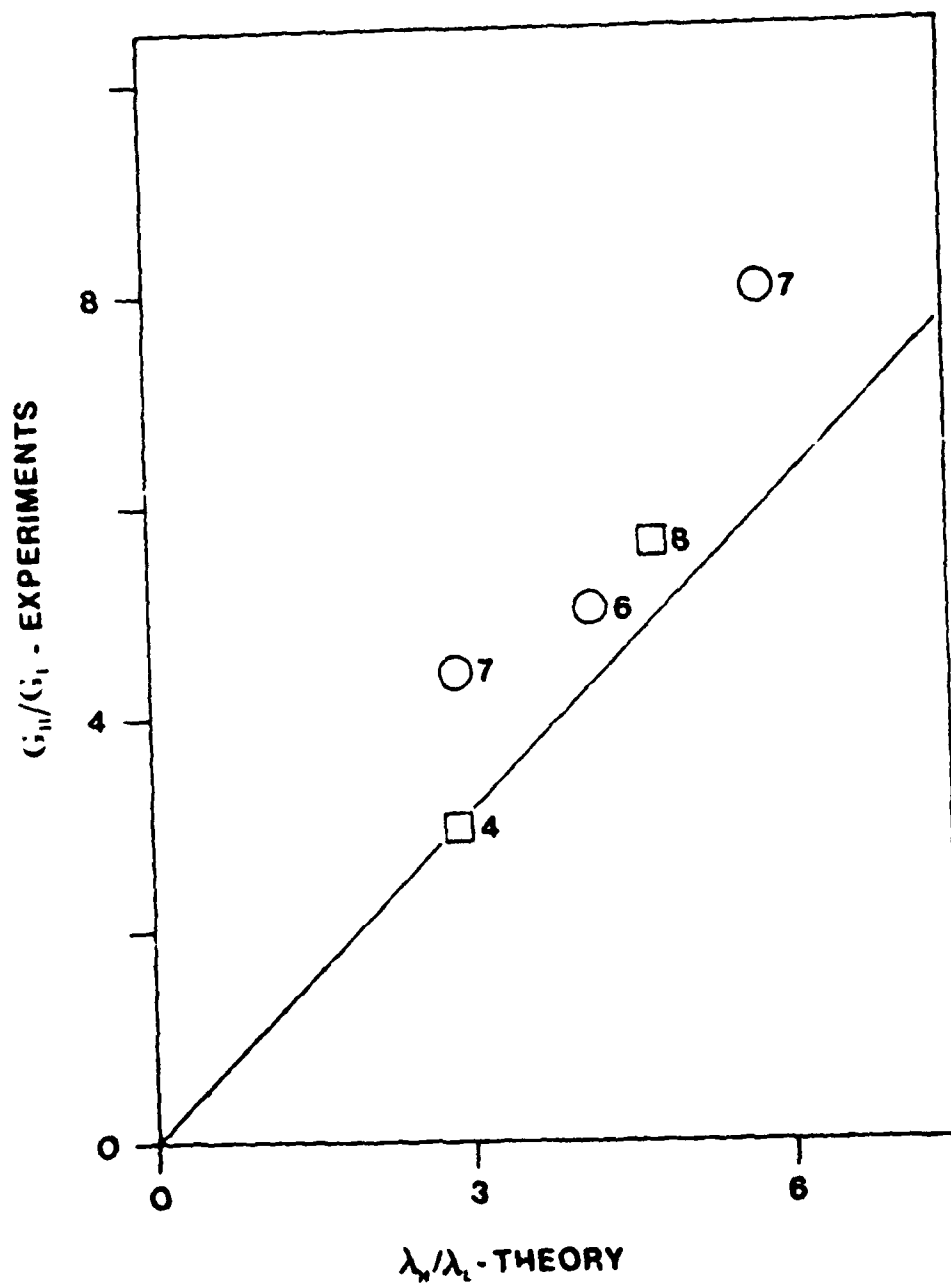


Fig. 8

[54] SYSTEM AND METHOD FOR SEPARATING GASES OF DIFFERING MASSES BY ENHANCED DIFFUSION PRODUCED BY TUNED OSCILLATIONS

[75] Inventors: Ulrich H. Kurzweg; Marc J. Jaeger, both of Gainesville, Fla.

[73] Assignee: University of Florida, Gainesville, Fla.

[21] Appl. No.: 2,268

[22] Filed: Jan. 12, 1987

[51] Int. Cl.<sup>4</sup> ..... B01D 53/22

[52] U.S. CL ..... 55/15; 55/17; 55/158

[58] Field of Search ..... 55/15-17, 55/158

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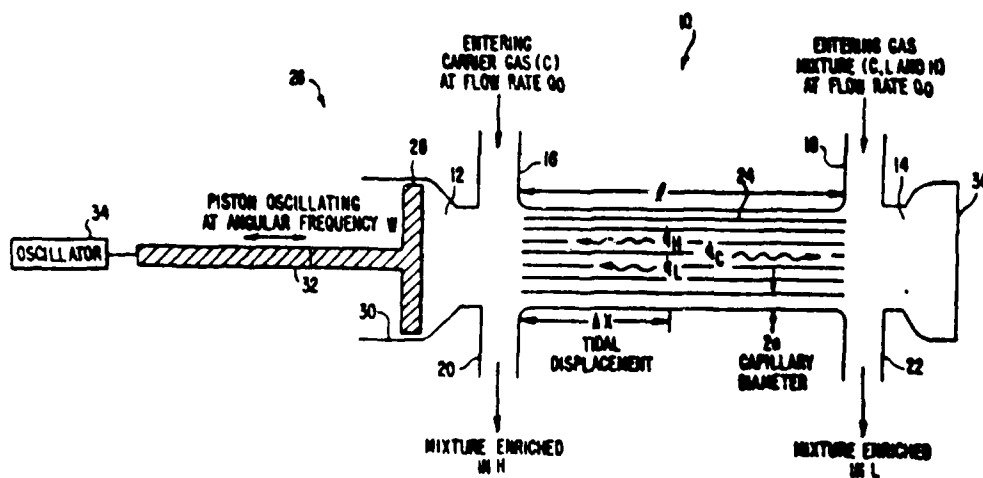
Attorney, Agent, or Firm—Dennis P. Clarke

[57]

ABSTRACT

A system for separating gases of differing masses by enhanced diffusion comprising a gas separating device which consists of a pair of gas reservoirs, at least one duct connecting the reservoirs and device for establishing oscillatory axial movement of gas within the duct. Each reservoir contains an inlet and outlet, the inlet being adapted for feeding a mixture of carrier gas and the mixture of gases to be separated and the inlet for the other reservoir being adapted for feeding a carrier gas into the other reservoir. One of the outlets is adapted for the withdrawal from one of the reservoirs of a mixture of carrier gas and a mixture of gases of differing masses which is enriched in the heavier gas component and the other outlet is adapted for withdrawal from the other of the reservoirs of a mixture of a carrier gas and a mixture of gases to be separated being enriched in the lighter gas component.

23 Claims, 5 Drawing Sheets



# SYSTEM AND METHOD FOR SEPARATING GASES OF DIFFERING MASSES BY ENHANCED DIFFUSION PRODUCED BY TUNED OSCILLATIONS

## BACKGROUND OF INVENTION

### 1. Field of the Invention

The present invention relates to a system and method for separating gases of differing masses by diffusion using tuned oscillations.

### 2. Prior Art

Paiva in U.S. Pat. No. 4,166,727 describes a method for separating gases of different masses by admitting a gas and the mixture of gases of different masses into one end of a duct so as to allow laminar flow of the admitted gases and collecting from the other end of the duct fractions of the mixture wherein the average relative concentration of one of the gases of different mass is higher than in the admitted mixture.

The differential diffusion method of Paiva suffers from several disadvantages. The patented method which attempts to increase separation rates in diffusion processes by using laminar axial convection in small diameter tubes working in conjunction with radial molecular diffusion requires switching devices for both the injection of gases into the system and for the removal of partially enriched and depleted mixtures.

Moreover, very long (about 10 meters) diffusion tubes are required by Paiva and the diffusion rates achieved are extremely low.

It is an object of the present invention to provide a differential diffusional method for separating gases of differing masses which are not subject to the above described disadvantages.

## SUMMARY OF THE INVENTION

The foregoing and other objects are realized by the present invention, one embodiment of which is a system for separating gases of differing masses from a mixture thereof by enhanced diffusion comprising at least one gas separating device comprising:

a pair of gas reservoirs, each having gas inlet and outlet means;

at least one duct connecting the gas reservoirs; and means for establishing oscillatory axial movement of gas within the at least one duct;

one of the inlet means being adapted for feeding into one of the reservoirs a mixture of a carrier gas plus the mixture of gases of differing masses;

the other of the inlet means being adapted for feeding a carrier gas into the other of the reservoirs;

one of the outlet means being adapted for the withdrawal from one of the reservoirs a mixture of carrier gas and a mixture of gases of differing masses enriched in the heavier gas component; and

the other of the outlet means being adapted for the withdrawal from the other of the reservoirs a mixture of carrier gas and a mixture of gases of differing masses enriched in the lighter gas component.

A further embodiment of the invention is a method for separating gases of differing masses from a gas mixture by enhanced diffusion comprising conducting at least one cycle of:

feeding a mixture of carrier gas plus the mixture of gases of differing masses into a gas reservoir which is connected by at least one duct to another gas reservoir;

feeding the carrier gas into the other gas reservoir; establishing oscillatory axial movement of gas within the at least one duct;

withdrawing from one of the gas reservoirs a mixture of carrier gas and a gas mixture of differing masses enriched in the heavier gas component thereof; and

withdrawing from the other of the reservoirs a mixture of carrier gas and a mixture of gases of differing masses enriched in the lighter component thereof.

## PRIOR ART STATEMENT

The following prior art is noted in conformance with the provisions of 37 CFR 1.97 and 1.98.

Paiva, U.S. Pat. No. 4,166,727 is described above.

Watson, *J. Fluid Mech.* Vol. 133, P. 233 (1983); Jaeger et al, *Phys. Fluids*, Vol. 26, p. 1380 (1983) examine the general problem of the enhanced dispersion of a contaminant in a carrier fluid when the mixture is oscillated axially within a tube in the presence of an axial concentration gradient. The publication presents an analytical study without experiments. Joshi et al, *J. Fluid Mech.*, Vol. 133, p. 245 (1983) verify Watson's predictions of enhanced gas dispersion by noting the experimental enhanced dispersion of methane gas in air.

It is important to realize that neither of the papers recognized that oscillations can be used for gas separation at high differential diffusion rates. The authors viewed the process as one of enhanced gas mixing.

Dreyer et al, *Naturforsch.* Vol. 23a, p. 498 (1968) examines the separation of liquid mixtures using low frequency oscillations (less than 1 Hz) of the fluid between parallel plates and pipes in the presence of a superimposed steady flow of water. The water is circulated through the system by an evaporation-condensation process. G. Dreyer and F. Lange hold a German patent, No. 54339 (1967) on this process. See also Lange et al, DWP No. 54339 and Lange et al, DWP No. 28316.

Taylor, *Proc. Roy. Soc.*, Vol. H 219, p. 186 (1983), is an original paper on enhanced dispersion of contaminants in steady viscous pipe flow. The reference, for the first time, shows that a combination of axial convection and radial diffusion can greatly enhance axial disposition of contaminants in liquids.

Hertz, *Z. Physik*, Vol. 19, p. 35 (1923); Lange et al, *Z. Naturwiss.*, Vol. 39, p. 765 (1930) and Maier, *J. Chem. Phys.*, Vol. 7, p. 854 (1939) disclose molecular diffusion methods to separate isotopes.

## BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a diagrammatical sectional view of an embodiment of a diffusion cell system according to the invention.

FIG. 2 is a diagrammatical sectional view of a carrier gas condenser for the system of the invention.

FIG. 3 is a diagrammatical sectional view of a cascade system of the invention.

FIG. 4 is a graphical depiction of the results of a method according to the invention.

FIG. 5 is a further graphical depiction of the results of a method according to the invention.

## DETAILED DESCRIPTION OF THE INVENTION

The present invention is predicated on the discovery that mixtures of gases of differing masses can be separated at high differential flow rates by an enhanced diffusion technique involving the oscillation of gases

within one or more ducts or capillaries in the presence of axial concentration gradients.

Referring to FIG. 1, a gas separation device 10 in accordance with the invention includes a pair of gas reservoirs 12 and 14, equipped with gas inlets 16 and 18, respectively, and gas outlets 20 and 22, respectively.

The two reservoirs 12 and 14 are connected by at least one, and preferably a plurality, of ducts 24. The ducts 24 preferably have a diameter of from about 1 mm to about 4 mm.

The reservoirs 12 and 14 and the ducts 24 are adapted to receive and hold and transfer gas, respectively.

Acting on gas between the reservoirs 12 and 14, in this example, within reservoir 12 is an oscillatory displacement device 26. As a representative example, the oscillatory displacement device 26 comprises a driving piston 28 reciprocating within a cylindrical bore 30 in direct communication with the reservoir 12 and driven via a shaft 32 by an external mechanical oscillator 34. Typically, the driving piston 28 oscillates sinusoidally, however, any suitable oscillation wave form may be used.

Since the gas is substantially incompressible, in order to keep the system entirely filled at all times with no voids or dilutions of the gas mixture, a displacement accommodating device 36 is connected to reservoir 14. In the example shown, the displacement accommodating device comprises what may be viewed as a passive resilient member, preferably a membrane.

With this arrangement, as the piston 28 oscillates, gas moves alternately axially in opposite directions within the ducts 24. The displacement accommodating member 36, due to its resilient nature, returns energy to the driving piston 28. The oscillation amplitude is selected such that the extent of gas movement within the ducts 24 is less than one-half of the length of the ducts.

Referring to FIG. 2, a typical condenser 100 is shown which is capable of partially removing the carrier gas from gas mixtures entering therein through inlets 102 by having it condense along condenser plates 104, cooled by a coolant entering via inlet 106 which exits the system via outlet 108 and collecting in liquid form 110 at the bottom of the condenser. The condensed liquid may be withdrawn via outlet 112. The resultant gas mixture depleted in carrier gas is withdrawn by outlet 114 at the condenser tank bottom. This condenser could also be replaced by a chemical absorption system which can remove the carrier gas as effectively as a condenser.

Referring to FIG. 3, a standard Hertz cascade cycle 200 is shown which can be used to produce an essentially complete separation of the gas components of a mixture. The cycle consists of three basic elements: diffusion tubes 10, condensers 100 and pumps 202. The gas mixture to be separated enters the system via injection port 204. Carrier gas enters via ports 206. In the system shown the light gas component diffuses faster down the diffusion cells 10 from right to left than does the heavy gas component. As a result the mixture exiting the left of each diffusion cell is enriched with the lighter species, while the mixture exiting the right side of the diffusion cell is enriched with the heavier molecules. The function of the condenser is to remove carrier gas periodically, while the pumps insure that there is no net convection flowing along the diffusion cells and that enriched gas mixtures are moved to the next stage.

It is believed that the principles, operation and exemplary embodiments of the invention, will all be under-

stood from the foregoing. Following is a detailed description of an embodiment of a method of the invention.

Referring again to FIG. 1, the gas at the injection port 16 is essentially 100% carrier gas (C) while the mixture at the injection port 18 is a gas mixture composed of the light (L) and heavy (H) molecules to be separated together with a small amount of carrier gas. Preferably, the same amount of continuous flow which enters the injection ports is withdrawn through ports 20 and 22 so that there is no convection flow along the tube bundle. The only gas exchange possible along the tubes is that produced by diffusion. If the piston oscillator, 26 is actuated to produce an axial periodic motion of the gas within the tube bundle, the effective axial diffusion can be made orders of magnitude larger than in the absence of oscillations. If the analytical results for such an enhanced diffusion process are examined it becomes clear that mass additives to a carrier fluid should diffuse at different rates down the tubes. The differential rate of migration depends on many parameters and, the diffusional flow rate of the light gas component (L) will be largest at or near the tuning point  $\alpha^2 f = (1/4) D_m$ , where  $\alpha$  is the individual tube radius,  $f$  the oscillator frequency and  $D_m$  the molecular diffusion coefficient of the light gas component under consideration. Typically, the frequency of oscillations will lie in the range of  $f = 5$  Hz to  $f = 30$  Hz. The diffusional flow of H can be either larger or smaller than that for L depending on the choice of tube radius and oscillator frequency. There is even a point for which no separation occurs. Very good separation will be accomplished when running at an oscillator frequency tuned for the L component. At the same time separation can also be achieved at smaller  $\alpha = \sqrt{2\pi f / \nu}$ , where  $\nu$  is the kinematic viscosity of the carrier gas. In this latter case the H molecules will actually diffuse faster down the tubes than does the L gas component. Experimental results, obtained with an apparatus similar to that shown in FIG. 1, after replacing the inlet 18 and exit 22 by a 20 liter box, have confirmed the ability of the present oscillatory approach to separate gas components. Using oxygen as the carrier gas, with about 10% each of helium and carbon dioxide added as the components to be separated, gave separation factors of  $p = 0.6$  at  $\alpha = 2$ . See Eq. (3) below for the definition of  $p$ .

#### EXAMPLE

Referring to FIG. 1, pure carrier gas (C), is injected into reservoir 12 through inlet 16. At inlet 18, a mixture of carrier gas and the mixture of light (L) and heavy (H) gases are injected into reservoir 14. In reservoir 12, the gas consists of almost pure carrier gas with a small amount of the L and H components which have diffused from reservoir 14 side of the capillaries 24. The injection flows on both sides are continuous and held at the same constant value of  $Q_0$ , as are the withdrawal rates through outlets 20 and 22. The piston oscillator 26 sets the gases within the capillaries into sinusoidal axial motion at angular frequency but the tidal displacement  $\Delta x$  is always kept sufficiently small so that a condition of zero net convection between the capillary ends is maintained. The time averaged axial diffusion flow rate of the L and H species to be separated, under these conditions, is

$$J_{L,H} = (D_m / 4) \frac{(\Delta C)^2}{L} \quad (1)$$

with  $c_{LH}(1)$  and  $c_{LH}(2)$  representing the concentrations in reservoirs 14 and 12, respectively,  $l$  the capillary length,  $A_0$  the total capillary bundle cross-section and  $D_{eff}$  the effective diffusion coefficient. The corresponding differential diffusional flow rate, directly obtainable from Eq. (1), is

$$\Delta q = q_H - q_L = Q_0 N \left[ \frac{c_H(1)}{N + \Gamma} - \frac{c_L(1)}{N + 1} \right] / L \quad \text{where } N = \frac{(D_{eff} L^2 / A_0 Q_0)}{c_L(2) [c_L(1) - c_L(2)] \text{ and } \Gamma = \frac{(D_{eff} L / (D_{eff} H))}{(D_{eff} L / (D_{eff} H))} \quad (2)$$

Gas component separation will occur whenever  $\Gamma$  is different from unity, while the separation flow  $\Delta q$  may be either positive or negative depending upon the values of  $N$ ,  $\Gamma$  and  $c_{LH}(1)$ .

Corresponding to the above diffusion flow rates, the separation factor for the light species is

$$\rho_L = \frac{c_L(2) c_H(1)}{c_L(1) c_H(2)} = \frac{N + \Gamma}{N + 1} \quad (3)$$

This quantity has values greater than one when  $(D_{eff} L) > (D_{eff} H)$  and less than one when the inequality is reversed. No separation occurs when  $\rho_L = 1$ .

The value of the effective diffusion coefficient,  $D_{eff}$ , may be determined from existing theory for contaminant dispersion in pipes provided one makes the restriction that one is dealing with dilute mixtures where the flow hydrodynamics is essentially controlled by the properties of the carrier gas while the diffusion of the L and H additives into C is assumed to be independent of each other. Such an assumption is reasonable for gas mixtures where the carrier has large concentrations compared to the L and H components and leads, after neglecting direct axial diffusion and assuming laminar conditions, to the result

$$D_{eff} = v(\Delta x / a)^2 H(a, S) \quad (4)$$

where  $a$  is the radius of the individual capillaries,  $v$  the kinematic viscosity of the carrier,  $\Delta x$  the gas tidal displacement and

$$H(a, S) = \frac{F(a) - \frac{1}{\sqrt{S}} F(a\sqrt{S})}{\left| 1 + \frac{1}{a} F(a) \right|} \quad (5)$$

$$\frac{a S}{4(S^2 - 1)} \left[ \frac{F(a) - \frac{1}{\sqrt{S}} F(a\sqrt{S})}{\left| 1 + \frac{1}{a} F(a) \right|} \right]$$

with the complex function

$$F = F_R + i F_I = i \left[ \frac{\text{ber}' a + i \text{bei}' a}{\text{ber} a + i \text{bei} a} \right] \quad (6)$$

related to the Kelvin functions  $J_0(i^{3/2} a) = \text{ber} + i \text{bei}$  whose tabulation may be found in the NBS Handbook of Mathematical Functions. The function  $H(a, S)$  can be derived and has the advantage that derivatives no higher than the first of the Kelvin functions are required. The results of a computer evaluation of Eq. (5)

are given in FIG. 4 which depicts effective diffusion coefficients as a function of molecular diffusion coefficients for several different values of the Womersley number  $\alpha = a\sqrt{\omega}/v$ . The quantity  $S$  used in the abscissa represents a modified Schmidt number equal to the ratio of the kinematic viscosity of the carrier gas to the binary molecular diffusion coefficient  $D$  of the particular species diffusing into the carrier. The value of  $S$  for gas mixtures range from about  $S=0.1$  for light molecules diffusing into a low  $v$  carrier gas to about  $S=10$  for heavy molecules diffusing into a high  $v$  carrier. Using standard formulas [Bird et al, *Transport Phenomena* (John Wiley & Sons, N.Y., 1960) pp. 17, 303-305] for  $v$  and  $D$  based on critical temperature and pressure and molecular weight, one finds at 300° K., that  $S=0.2$  for He into O<sub>2</sub>,  $S=1.0$  for CO<sub>2</sub> into O<sub>2</sub> and  $S=1.6$  for SF<sub>6</sub> into O<sub>2</sub>. According to FIG. 4, good separation of gas components should be possible whenever the absolute value of  $\partial H / \partial S$  is large and values of  $S$  differ between the constituents to be separated. No separation will occur where  $\partial H / \partial S$  is zero or when  $S_L = S_H$ . As seen from Eq. (2), a maximum in the differential diffusion flow rate can be expected for  $N = \sqrt{\Gamma}$ , whenever  $c_L(1) = c_H(1)$ .

The following specific example was carried out using a system substantially identical to that shown in FIG. 1, except that reservoir 14 was replaced by a large 20 liter hermetically sealed box into which specified gas mixtures could be placed. Although this geometry allows only quasisteady separation to be carried out, it has the advantage that changes in box pressure can be used directly to insure that no convection flow exists in the diffusion cell during the course of an experiment. Oxygen was chosen as the carrier gas. The injection flow rate of the O<sub>2</sub> carrier at entry port 16 was maintained at  $Q_0 = 8$  cc/sec. Moreover, a suction pump (not shown) is located at exit port 20. The oscillation frequencies used in these experiments ranged from 2 to 27 Hz and the tidal displacements from 4 to 30 cm. Two sets of capillary tubes having  $a = 0.05$  cm,  $A = 2.8$  cm<sup>2</sup>,  $l = 34$  cm and  $a = 0.1$  cm,  $A = 2.8$  cm<sup>2</sup>,  $l = 86.5$  cm were used. In addition, a single rigid tube with  $a = 0.79$  cm,  $A = 2.0$  cm<sup>2</sup>,  $l = 140$  cm was also employed. In none of the runs was  $\Delta x$  made larger than  $\frac{1}{2}$  of the tube length in question. For the gas mixture to be separated combinations of CO<sub>2</sub>-He and CO<sub>2</sub>-SF<sub>6</sub> were chosen since these combinations have a large molecular weight difference and also their concentration is readily measurable with readily available equipment. Results of some 40 readings on gas separation with these gas mixtures are recorded in FIG. 5 as  $\rho_L$  versus  $N$ . FIG. 5 depicts separation factor as a function of effective diffusion coefficient. The triangles represent CO<sub>2</sub>-He mixtures and the circles, SF<sub>6</sub>-CO<sub>2</sub> mixtures. The average carrier concentration was  $C_c = 0.8$  or greater. The solid curves represent an evaluation of Eq. (3) for different values of  $\Gamma$ . The range of Womersley number used for both mixtures was from  $\alpha = 0.5$  to  $\alpha = 12$ . The results clearly show that partial separation has been achieved, with the He-CO<sub>2</sub> results clustering about an average value of  $\Gamma = 0.6$  and the CO<sub>2</sub>-SF<sub>6</sub> results around  $\Gamma = 1.2$ . A comparison with the analytical result given by Eq. (3) and plotted in FIG. 5 for several different values of  $\Gamma$  clearly shows that the experimental results are in good qualitative agreement with theoretical predictions but yield somewhat lower separation values, especially for He-CO<sub>2</sub> mixtures. Part of this discrepancy is believed due to the use of carrier

gas concentrations as low as  $c_L(1)=0.6$ , needed to keep the values of  $C_L(2)$  and  $c_H(2)$  above 0.01 but in violation of the assumption concerning dilute mixtures. For He-CO<sub>2</sub> mixtures the best separation was observed to occur near  $\alpha=1$  and no separation was found above  $\alpha=8$ . This is in agreement with the values for  $\partial H/\partial S$  shown in FIG. 4. The best separations for CO<sub>2</sub>-SF<sub>6</sub> mixtures occurred for  $\alpha>10$  and these were characterized by diffusion rates for CO<sub>2</sub> greater than those for SF<sub>6</sub>. A large reversal of this diffusion behaviour in CO<sub>2</sub>-SF<sub>6</sub> mixtures was not observed as  $\alpha$  was brought down to values as low as  $\alpha=0.5$ , although theory would have predicted such a reversal. A final interesting observation deals with the differential diffusion flow rate predicted from Eq. (2) by using measured values from the data shown in FIG. 5. Taking for example the point at  $pL=0.65$  and  $N=0.3$  for the He-CO<sub>2</sub> mixture, where  $\Gamma$  is 0.6 and for which the concentrations at 1 were  $c_L(1)=c_H(1)=0.2$ , one finds via Eq. (2), that  $\Delta\dot{q}=0.25$  cc/sec. This compares with the value of  $\Delta\dot{q}=-0.0026$  cc/sec which would be expected for the same mixture and conditions in the absence of oscillations in the same length tube. It is this ability of oscillations to increase the differential diffusion flow rates by several orders of magnitude which makes separation technique of the invention highly advantageous and efficient.

The above example shows that sinusoidal oscillations may be used to produce component separations of gas mixtures at much higher separation flow rates than in the absence of oscillations. Separation factors as low as  $pL=0.5$  were produced for He-CO<sub>2</sub> mixtures and as high as  $pL=1.5$  for SF<sub>6</sub>-CO<sub>2</sub> mixtures in a single path through one diffusion cell. To obtain essentially complete separation it would be necessary to conduct the process in cascade fashion in which several diffusion cells are coupled together as shown in FIG. 3. Also, there would be need for the periodic removal of carrier gas in the enriched and depleted portions of the gas mixtures leaving the exit ports of the individual diffusion cells by, for example, the condensers shown in FIGS. 2 and 3. Carrier gases such as steam or CO<sub>2</sub> would be likely candidates for practical applications of the separation process of the invention as these gases can be readily condensed or removed by chemical means. Finally, it should be pointed out that the present separation process works best for relatively large values of  $\Delta\chi$  (and hence of  $l$ ) and is not very effective as  $l$  becomes small, where direct axial molecular diffusion will dominate.

The oscillation approach of the present invention using tuned conditions yields diffusion flow rates much higher than possible by the steady-flow method described by Paiva. The effective diffusion coefficients, as described in column 5 of U.S. Pat. No. 4,166,727, are about 4 cm<sup>2</sup>/sec while those obtained herein are easily above 1000 cm<sup>2</sup>/sec and more. The oscillation approach is a continuous flow process not involving switching devices as necessary in the Paiva diffusion method.

The oscillation approach uses short capillary tubes (less than 1 meter long) at oscillating frequencies greater than the 5 Hz. Paiva requires tubes of 10 meters (~33 ft.) length.

We claim:

1. A method for separating gases of differing masses from a gas mixture by diffusion comprising conducting at least one cycle of:

feeding a mixture of carrier gas and said mixture of gases of differing masses into a gas reservoir which

is connected by at least one duct to another gas reservoir, the space defined by said gas reservoirs and said at least one duct being further undivided; feeding said carrier gas into said other gas reservoir; establishing oscillatory axial movement of gas within said at least one duct;

withdrawing from one of said gas reservoirs a mixture of carrier gas and a gas mixture of differing masses enriched in the heavier gas component thereof; and

withdrawing from the other of said gas reservoirs a mixture of carrier gas and a mixture of gases of differing masses enriched in the lighter gas component thereof.

2. The method of claim 1 wherein said reservoirs are corrected by a plurality of ducts.

3. The method of claim 1 wherein said oscillatory axial movement is sinusoidal and tuned to maximize diffusion of one of the components of the gas mixture.

4. The method of claim 1 comprising removing at least a portion of carrier gas from at least one of said withdrawn gas mixtures.

5. The method of claim 4 wherein said carrier gas is removed by condensation.

6. The method of claim 1 comprising conducting multiple said cycles in cascade fashion wherein at least one of said mixtures of carrier gas and gas mixture enriched in one of said heavier or lighter components thereof withdrawn from a gas reservoir in a previous cycle is fed into a gas reservoir in a subsequent cycle whereby each succeeding withdrawn mixture is more enriched in said lighter or heavier component than the previously withdrawn mixture.

7. The method of claim 6 wherein at least a portion of carrier gas is removed from at least one of said withdrawn gas mixture.

8. The method of claim 7 wherein said carrier gas is removed by condensation.

9. The method of claim 1 wherein said gas is alternately moved axially in opposite directions within said at least one duct, that extent of movement therewithin being less than the length thereof.

10. The method of claim 1 wherein the rates of feeding and withdrawal of said gases and gas mixtures are equal.

11. The method of claim 1 wherein the tidal displacement in said at least one duct is maintained sufficiently small so as to maintain a condition of zero net convection between the ends of said at least one duct.

12. A system for separating gases of differing masses from a mixture thereof by enhanced diffusion comprising at least one gas separating device comprising:

a pair of gas reservoirs, each having gas inlet and outlet means;

at least one duct connecting said gas reservoirs, the space defined by said pair of gas reservoirs and said at least one duct being further undivided; and means for establishing oscillatory axial movement of gas within said at least one duct;

one of said inlet means being adapted for feeding into one of said reservoirs a mixture of a carrier gas and said mixture of gases of differing masses;

the other of said inlet means being adapted for feeding a carrier gas into the other of said reservoirs;

one of said outlet means being adapted for the withdrawal from one of said reservoirs a mixture of carrier gas and a mixture of gases of differing masses enriched in the heavier gas component; and



the other of said outlet means being adapted for the withdrawal from the other of said reservoirs a mixture of carrier gas and a mixture of gases of differing masses enriched in the lighter gas component.

13. The gas separation system of claim 12 comprising a plurality of said ducts connecting said gas reservoirs.

14. The gas separation system of claim 12 wherein said means for establishing oscillatory axial movement establishes a sinusoidal movement.

15. The gas separation system of claim 12 wherein at least one of said outlet means is connected to means for removing at least a portion of said carrier gas from said withdrawn gas mixture.

16. The gas separation system of claim 15 wherein said means for removing carrier gas is a condenser.

17. The gas separation system of claim 12 comprising multiple said gas separating devices connected in cascade fashion such that at least one of said mixtures of carrier gas and gas mixture enriched in one of said lighter or heavier components thereof withdrawn from a reservoir of a previous gas separating device is fed into a reservoir of a subsequent gas separating device, whereby each succeeding withdrawn mixture is more

enriched in said component than the previously withdrawn mixture.

18. The gas separation system of claim 17 wherein said outlet means of at least one of said multiple gas separating devices is connected to means for removing at least a portion of said carrier gas from said withdrawn gas mixture.

19. The gas separation system of claim 18 wherein said means for removing said carrier gas is a condenser.

20. The gas separation system of claim 12 wherein said means for establishing oscillatory axial movement of gas within said at least one duct comprises an oscillatory displacement device acting on gas within one of said reservoirs for causing gas to alternately move axially in opposite directions within said at least one duct, the extent of gas movement therewithin being less than the length thereof.

21. The gas separation system of claim 20 wherein the other of said reservoirs contains a displacement accommodating device acted on by the gas contained therein.

22. The gas separation system of claim 21 wherein said displacement accommodating device comprises a passive resilient member.

23. The gas separation system of claim 22 wherein said passive resilient member is a membrane.

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FIG. 1.

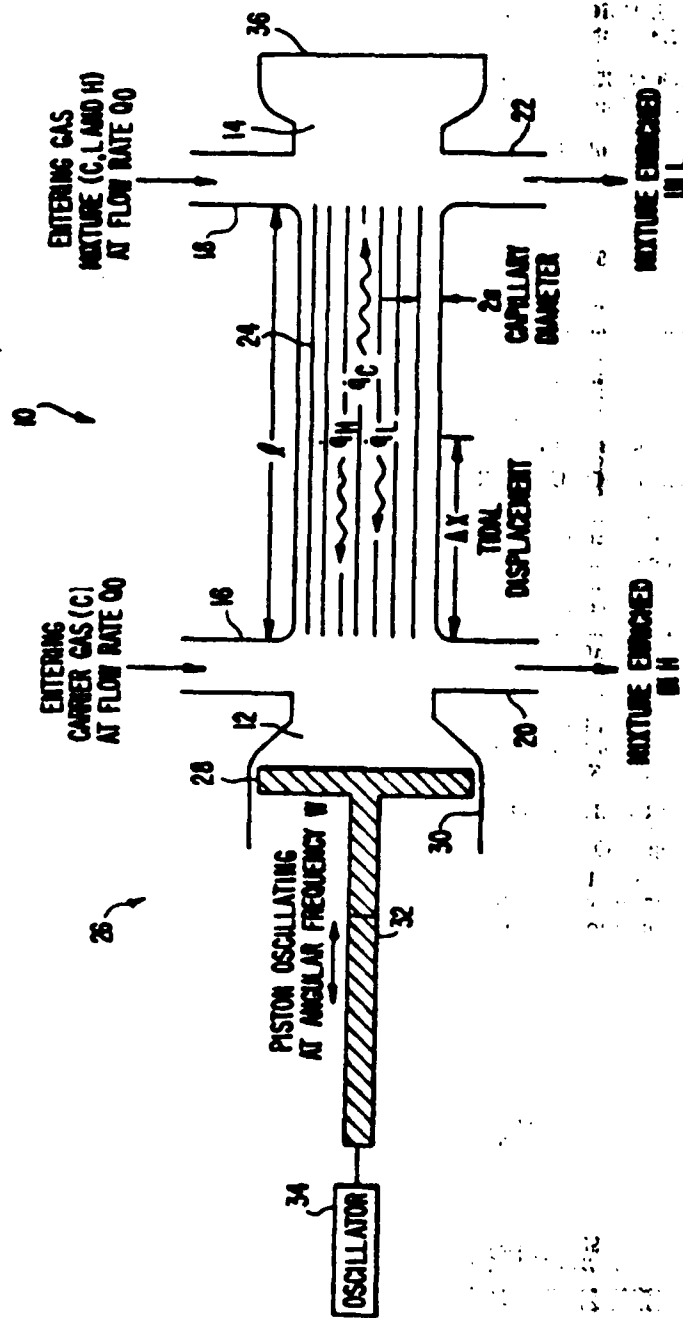


FIG. 2.

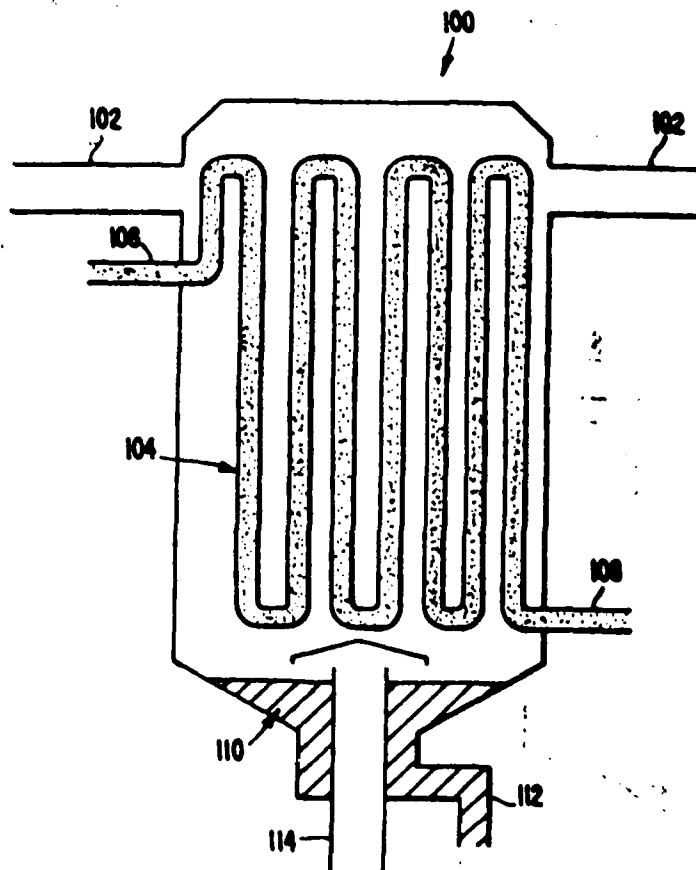


FIG. 3.

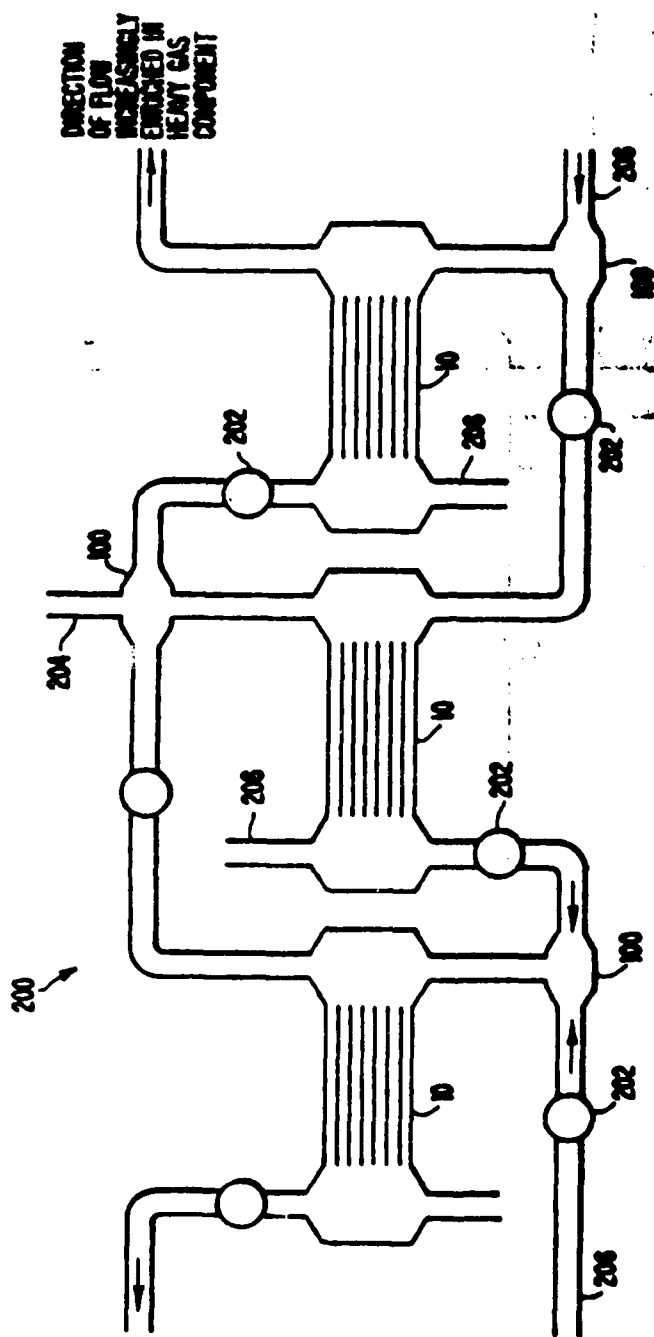


FIG. 4.

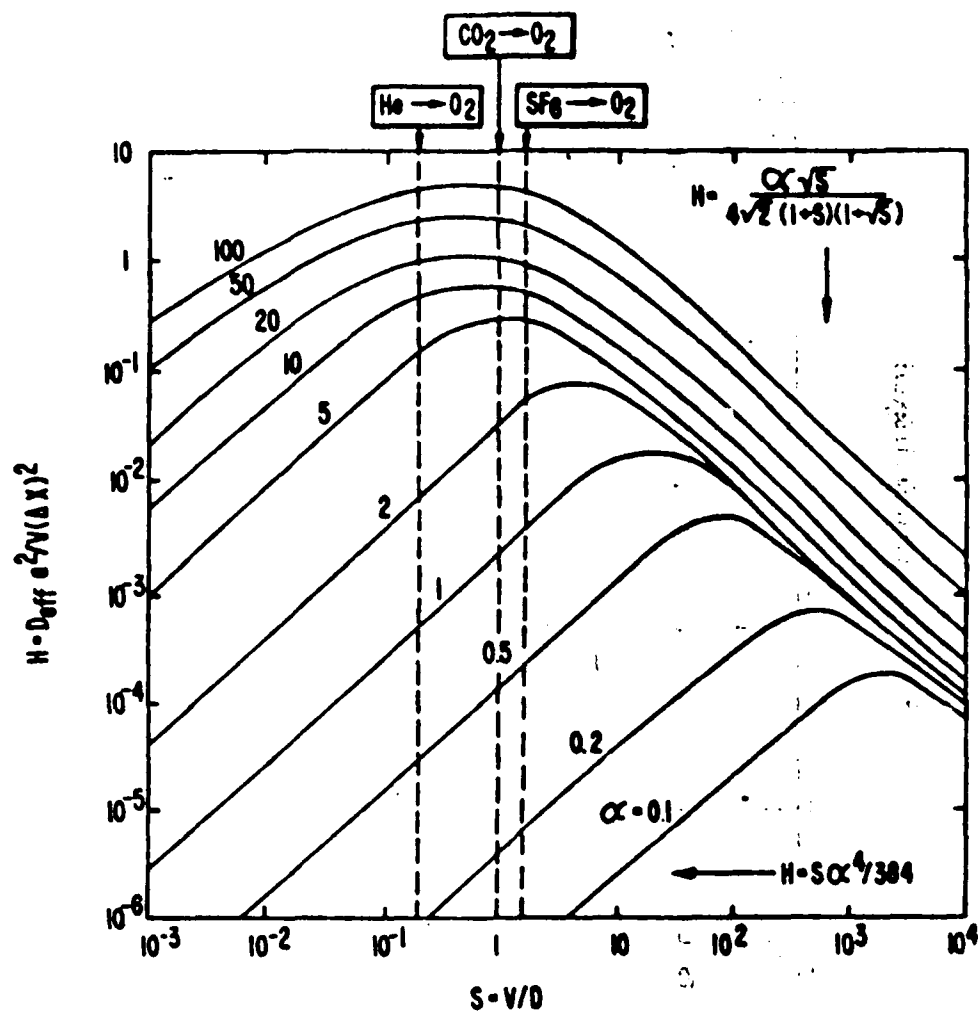


FIG. 5.

